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SOME AMPLIFICATION BY RELAXATION PROCESSES IN NITROGEN
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PHYSICAL ACOUSTICS RESEARCH LAB F D SHIELDS ET AL.

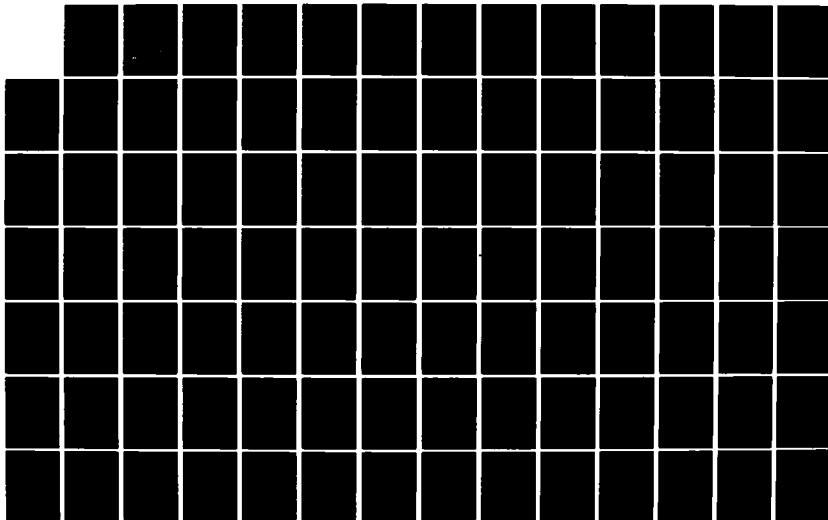
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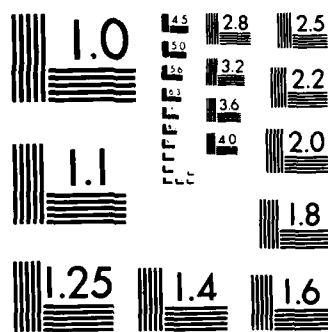
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SOUND AMPLIFICATION BY RELAXATION PROCESSES
IN NITROGEN AND MIXTURES OF NITROGEN
WITH HELIUM AND HYDROGEN

F. Douglas Shields and Dadang Iskandar

PARGUM Report 85-01

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THE UNIVERSITY OF MISSISSIPPI
PHYSICAL ACOUSTICS RESEARCH GROUP
DEPARTMENT OF PHYSICS AND ASTRONOMY

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compared to determine the gain and temperature of the gas. In pure nitrogen and mixtures of nitrogen and helium, in which the vibrational relaxation time was much longer than the sound period, gain was not observed. Gain was observed in three cases: 50% He - 50% N₂ at 40 Torr, 10% H₂ - 90% N₂ at 40 and 20 Torr. In the first two mixtures it was possible to observe the change in gain with time as the vibrational temperature decayed away. (Right for Safford keywords in file)

Preface

This report is a reproduction of the Ph.D. dissertation prepared by Dadang Iskandar under the direction of F. Douglas Shields. Certain pages in the dissertation required by the University but not contributing to the technical content here have been omitted. The dissertation was accepted by the faculty of the Department of Physics and Astronomy in February 1985.

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SOUND AMPLIFICATION BY RELAXATION PROCESSES IN NITROGEN
AND MIXTURES OF NITROGEN WITH HELIUM AND HYDROGEN

BY

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B.S., Gajah Mada University, 1968
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CHAPTER I

INTRODUCTION

There have been a number of theoretical studies of the amplification of sound propagating through gases under a variety of non-equilibrium conditions. Ingard and others¹⁻³ have predicted the amplification of sound propagating in a weakly ionized gas. Gilbert and others⁴⁻⁶ predicted the amplification of sound propagating in reacting systems. De Plomb,⁷ Schulz,⁸ Bauer and Bass,⁹ and Shields¹⁰ predicted the amplification of sound propagating in gases subjected to external energy input (energy pumping). In the latter cases two different mechanisms leading to the amplification of sound have been proposed: selective absorption of the pumping energy and relaxation processes of internal modes. The sound amplification due to the relaxation processes results from the fact that the vibrational energy transition rate is pressure and temperature dependent. The increase in pressure and temperature results in an increase in the transition rate, which in turn results in a further increase in temperature (here, temperature means translational temperature). Thus, in the presence of the sound wave, the vibrational relaxing energy will be selectively transferred into the hot part of the sound cycles. Experimental study to verify these theories is still limited. Recently Bass and Detsch¹¹ observed the amplification by gas phase reactions in Cl-H₂-Ar and Cl-H₂-SF₆ mixtures. In this study, the amplification of sound by vibrational relaxation processes is of primary interest. In order to

separate the effect due to relaxation from that due to pumping, the pumping mechanism is terminated before the sound wave is introduced into the gas.

When energy is pumped into the gas and then the pumping process is terminated, a metastable state with an overpopulation of excited vibrational states results. Relaxation to equilibrium occurs due to vibrational-translational (v-t) energy transfer, or, in the case of polyatomic gases due to vibrational-vibrational (v-v), followed by the v-t energy transfer. The amplification of sound wave should occur when it is propagated through the gas during the relaxation process. In this case, however, the sound period must be short compared to the relaxation time. Shields¹⁰ has considered three typical situations in which it is experimentally feasible to observe the amplification of sound. The three situations are:

1. Metastable state with a single long lived relaxing mode;
2. Metastable state with fast v-v, slow v-t exchanges; and
3. Metastable state with slow v-v, fast v-t exchanges.

The first case can be produced in most diatomic gases. Diatomic gases such as N_2 , O_2 , Cl_2 , and CO have a single vibrational mode with a long relaxation time. After energy is pumped into excited vibrational levels a metastable state persists for a period of time longer than the period of low-frequency sound. The last two cases can be produced in polyatomic gases or some mixtures. Polyatomic molecules possess several vibrational

modes which, in principle, can relax independently. Most polyatomic gases, however, show a single relaxation time.^{12,13} This is due to the rapid v-v energy transfer which maintains continuous equilibrium between modes. The amplification of sound propagating in such gases could be observed if the sound period is short compared to the v-t relaxation time. In some gases the v-v transfer may be slower than the v-t transfer.^{12,14} In this situation the sound period must be short compared to the v-v relaxation time. As examples of the last two cases, Shields¹⁰ calculated the expected sound amplification in CD_4 and in a $\text{N}_2\text{-He-CO}_2$ mixture.

In this investigation the sound wave is propagated through a vibrationally excited pure nitrogen gas and mixtures of nitrogen with helium and hydrogen. The overpopulation of vibrationally excited states is produced by an electrical discharge. A sound pulse is generated in the gas by the sudden increase in gas temperature accompanying the discharge. In this situation the sound pulse propagates immediately after the metastable state has been established, so that the requirement discussed above is satisfied. Nitrogen gas was chosen for the following reasons:

1. Nitrogen molecules have long relaxation times of the order of seconds at atmospheric pressure; and
2. Studies have shown that a large percentage of the energy from the electrical discharge will wind up in vibrational energy of nitrogen molecules.¹⁵

Nitrogen-helium and nitrogen-hydrogen mixtures were used to see the effects of changing the vibrational relaxation time and the thermal conduction relaxation time. The addition of helium and hydrogen molecules drastically decreases the relaxation time of nitrogen.¹⁶⁻¹⁹

The sound absorption and velocity in the gas were determined by observing the decay and the transit time of the compressional pulse that reflected back and forth in the gas. To determine the variation of the absorption and velocity (the translational temperature was determined from the velocity) with frequency and time, the pulse was Fourier analyzed after traveling various distances in the gas sample, and the absorption and velocity were determined for each frequency component.

CHAPTER II

THEORY

CALCULATION OF THE SOUND ABSORPTION AND VELOCITY IN AN EQUILIBRIUM GAS

As discussed in the introduction, the amplification of sound by relaxation processes results from the preferential heating of the compression part of the sound wave from the vibrational relaxation energy. However, the gain in energy will be countered by losses due to a number of effects. To observe the effects of the excited vibrational modes on the sound propagation it is therefore necessary to determine the absorption and velocity in the unexcited gases.

The most important effects leading to the sound absorption are shear viscosity, heat conduction, loss to the wall of the tube, and thermal relaxation. The absorption resulting from viscosity and thermal conduction is termed classical absorption. The classical equation for sound absorption and velocity in a tube was first developed by Kirchhoff.²¹ Beginning with the conservation equations for mass, energy, and momentum, he obtained an algebraic equation for the propagation constant for radially symmetric waves by approximating Bessel functions with a one term expansion. In this derivation he assumed that the particle velocity and the sound temperature were zero at the tube wall. Shields²² has solved for the absorption and velocity numerically and included the skin velocity and the temperature jump at the tube wall. In this study the

sound absorption and velocity in the unexcited gas were calculated at various frequencies and temperatures using the numerical solution developed by Shields.²³ The computer program for this numerical solution allows for calculation of viscous and thermal losses both in the body of the gas and at the tube wall and for relaxation effects. For this purpose the viscosity of gases used in this study were computed over the temperature range from 290°K to 350°K using formulae and potential parameters of the gas given in Reference 23. The specific heat for pure gases were taken from the Table of Thermal Properties of Gases.²⁴ The specific heat of mixtures were obtained by taking a weighted average of the pure component specific heats. The relaxation times were taken from the work of Frey, et al.¹⁷ and Audibert, et al.¹⁷. The momentum accommodation coefficient and the energy accommodation coefficient were set equal to 1.0 and 0.8 respectively. The values are approximate but have little effect on the calculated absorption and velocity. The physical properties of gases under study are given in Appendix B. To check the accuracy of this calculation, measurements on stable pure nitrogen gas and helium-nitrogen mixtures were made at various pressures and frequencies. The experimental method and results are discussed in Chapters III and IV.

CALCULATION OF THE ABSORPTION AND VELOCITY IN THE GAS WITH A METASTABLE STATE

In this section the equations leading to the amplification of sound are developed, and the numerical solutions to the resulting equations for

several gases are presented. Assuming that the attenuation and gain rates are additive, the amplification factor may be determined separately.

The relaxation absorption (or amplification as the case may be) and velocity can be obtained from the propagation constant for the acoustic wave. Using the continuity equations for mass, momentum and energy and neglecting the internal friction and thermal conduction, the propagation constant is given by

$$k = \sqrt{M/RT} \sqrt{\partial \rho / \partial P}. \quad (1)$$

For an ideal gas Equation 1 becomes

$$k = \omega \sqrt{M/RT} \sqrt{C_v / C_p}, \quad (2)$$

where ω is the angular frequency, M is the molecular weight, R is the gas constant, T is the absolute temperature, and C_v and C_p are the specific heats at constant volume and pressure. For a relaxing gas C_p and C_v are complex and frequency dependent. Writing k in Equation 2 in the complex form

$$k = k_1 - jk_2, \quad (3)$$

where $j^2 = -1$, the sound absorption (α_s) and the velocity (v_s) are given by

$$\alpha_s = k_2; \quad v_s = \frac{\omega}{k_1}. \quad (4)$$

Sound amplification results if k_2 is negative. The problem of calculating the absorption and velocity is now reduced to finding the real and imaginary part of C_p and C_v which can be derived from the reaction equation that gives the time rate of change of energy in a vibrational mode. The following was adapted from the work of Shields.¹⁰

When nitrogen gas is subjected to an electrical discharge most of the energy deposited in the gas will wind up in the vibrational energy of molecules,¹⁵ leaving the gas in the state with an overpopulation of excited vibrational states. Due to molecular collisions some of the vibrational energy will be converted back to the translational energy by v-v and/or v-t exchanges. A sound wave propagated during this transition process will cause small sinusoidal perturbations in the slowly decaying values of temperature and pressure. Assuming that the normal modes of molecular vibration can be treated as harmonic oscillators, and that resonant exchange of energy between levels within a single mode rapidly establishes a Boltzman distribution, the rate of change of energy in the ath vibrational mode can be written as²⁵

$$\dot{E}_a = \sum_{i,j} i h \nu_a P \sum_{\alpha=0}^{\infty} \sum_{\beta=0}^{\infty} [k^{\alpha \rightarrow \alpha+i; \beta \rightarrow \beta+j}(a,b) \frac{n_{\alpha}(a)n_{\beta}(b)}{N_b} - k^{\alpha+i \rightarrow \alpha, \beta+j \rightarrow \beta}(a,b) \frac{n_{\alpha+i}(a)n_{\beta+j}(b)}{N_b}] \quad (5)$$

where α and β refer to the vibrational quantum number of modes a and b respectively, P is the pressure, N_b is the total number of molecules carrying mode b , n_{α} and n_{β} are the numbers of molecules occupying α and β levels in modes a and b respectively, h is the Planck's constant, ν_a is

carrying mode b, n_α and n_β are the numbers of molecules occupying α and β levels in modes a and b respectively, h is the Planck's constant, ν_a is the fundamental vibrational frequency of mode a, and $k^{\alpha \rightarrow \alpha+i, \beta \rightarrow \beta+i}$ is the composition average rate constant in the dimension (time pressure)⁻¹ for the particular process in which α changes to $\alpha+i$ and β to $\beta+i$. The summation over α and β gives a net rate constant for transitions where mode a gains i quanta and mode b gains i quanta.

Equation 5 assumes that mode a is exchanging energy with mode b and with translation ($j=0$, $n/N_b=1$). Modes a and b can be in the same molecules, or, in the case of mixture, in different molecules and may have different temperatures. Assumptions that the normal vibrations can be treated as harmonic oscillators and a Boltzmann distribution is maintained allows us to write the following relations

$$k^{\alpha \rightarrow \alpha+i, \beta \rightarrow \beta+i} = \{[\alpha(\alpha-1)(\alpha-2)\dots(\alpha-i+1)] \times [(\beta+i)(\beta+i-1)\dots(\beta+1)]/i!j!\} \times k^{1 \rightarrow 0, 0 \rightarrow i}, \quad (6)$$

and

$$n_\alpha(a) = n_0(a) e^{-h\nu_a/kT_a} = N_a (1 - e^{-h\nu_a/kT_a}) e^{-h\nu_a/kT_a}, \quad (7)$$

where N_a is the number of molecules with mode a. Using Equations 6 and 7 and the relation

$$(\alpha+i)(\alpha+i-1)\dots(\alpha+1)x^i = \frac{d^i}{dx^i} \left(\frac{1}{1-x} \right) = \frac{i!}{(1+x)^{i+1}} \quad (8)$$

Equation 5 can be written as

$$\dot{E}_a = \sum_{i,j} i N_a h \nu_a P[k^{i \rightarrow 0, 0 \rightarrow i}_{(a,b)} e^{-i\theta_a/T_a} - k^{0 \rightarrow i, i \rightarrow 0}_{(a,b)} e^{-i\theta_b/T_b}] / ([\chi_a(T_a)]^i [\chi_b(T_b)]^i), \quad (9)$$

where $\theta_a = \frac{h\nu_a}{k}$; $\chi_a(T_a) = 1 - e^{-\theta_a/T_a}$. Here the principle of detailed balancing, $k^{i \rightarrow i+1} = k^{j+1 \rightarrow i} e^{-i\theta/T}$, has been used. Equation 9 can now be specialized to the three situations discussed in the introduction.

Most diatomic gases have only a single vibrational mode with a long relaxation time. In this situation, the vibrational mode, say mode a, communicates only with the translational mode so $j=0$ and $i=1$. Equation 9 then becomes, after some manipulations,

$$\dot{E}_a = -Pk^{10} \chi_a(T_a) [E_a(T_a) - E_a(T)] \quad (10)$$

where $E_a(T_a) = \frac{N_a k \theta_a \exp(-\theta_a/T_a)}{\chi_a(T_a)}$ and $E_a(T) = \frac{N_a k \theta_a \exp(-\theta_a/T)}{\chi_a(T)}$. When a sound wave is introduced \dot{E}_a will depart from its steady state value. Let $\Delta \dot{E}_a$ be the change in \dot{E}_a due to the presence of the sound wave. $\Delta \dot{E}_a$ can be obtained by differentiating Equation 9 with respect to the translational temperature T. The result is given by

$$\Delta \dot{E}_a = \dot{E}_a^0 \left\{ \left[\frac{1}{P} \frac{dP}{dT} + \frac{1}{k^{10}} \frac{dk^{10}}{dT} - \frac{\theta_a \exp(\theta_a/T)}{T^2 \chi_a(T)} \right] \Delta T - \frac{\Delta E_a(T_a) - \Delta E_a(T)}{\tau \dot{E}_a^0} \right\}. \quad (11)$$

Here only the first order terms of the sound variable are maintained. \dot{E}_a^0 , T_a , and T represent values without the sound wave. Introducing the sinusoidal time dependence and setting $\Delta E_a(T_a) = C_a(T_a) \Delta T_a$, $\Delta E(T) = C_a(T) \Delta T$ and $\dot{E}_a^0 = -\frac{E_a(T_a) - E_a(T)}{\tau}$, equation 11 becomes

$$(1+j\omega\tau)\frac{\Delta T_a}{\Delta T} = \left[\frac{\dot{E}_a^0 \tau}{C_a(T_a)} \right] \times \left[\frac{1}{P} \frac{dP}{dT} + \frac{d(\ln k^{10}(a))}{dT} - \frac{\theta_a e^{-\theta_a/T}}{T^2 \chi_a(T)} \right] - \frac{C_a(T)}{C_a(T_a)} \quad (12)$$

In the case in which the gas is in equilibrium (the gas has not been pumped) $T_a = T$ and $\dot{E}_a^0 = 0$. Equation 12 becomes

$$\frac{\Delta T_a}{\Delta T} = (1+j\omega\tau)^{-1} \quad (13)$$

and the frequency dependent specific heat is given by

$$C_v(\omega) = C_\infty + C_a \frac{\Delta T_a}{\Delta T} = C_\infty + \frac{C_a}{1+j\omega\tau}; \quad C_p(\omega) = C_v(\omega) + R \quad (14)$$

where C_∞ is the frequency independent part of the specific heat. When the gas has been pumped the vibrational temperature T_a is not equal to the translational temperature T and \dot{E}_a^0 is not zero. The frequency dependent specific heat is now given by

$$C_v(\omega) = C_\infty + C'_a (1+j\omega\tau)^{-1}; \quad C_p(\omega) = C_v(\omega) + R - \frac{\dot{E}_a^0 \tau}{T(1+j\omega\tau)}, \quad (15)$$

where

$$C'_a = (\dot{E}_a^0 \tau) \left[\frac{1}{T} + \left[\frac{d(\ln k^{10}(a))}{dT} - \frac{\theta_a e^{-\theta_a/T}}{T_a^2 \chi_a(T)} \right] \right] + C_a(T_a) \quad (16)$$

and R is the universal gas constant. The extra term, $\dot{E}_a^0 \tau / T(1+j\omega\tau)$, the second equation of Equation 14 results from the fact that, at constant pressure, $\frac{\partial P}{\partial T} = 0$. Amplification will occur if C'_a becomes negative. This is the case when $T_a - T$ is a few tens of degrees and $d(\ln k^{10})/dT$ (with dimension K^{-1}) is of the order of a few one hundreds. However, the amplification may be small and too difficult to observe. Using Equations

2,4,15, and 16, Shields¹⁰ has calculated the amplification factor for several diatomic gases. The results are shown in Table 1.

TABLE 1

Sound amplification in dB per wavelength for typical diatomic gases at 300°K. $d(\ln K^{10})/dT$ is set equal to 0.01, and $\omega\tau = 100$.

GAS	(cm) ⁻¹	Amplification factor (dB/λ)					
		T _a =500°K	1000°K	2000°K	3000°K	4000°K	6000°K
N ₂	2331	0.0027	0.082	0.52	1.1	1.7	2.9
CO	2143	0.0042	0.10	0.56	1.2	1.8	3.0
O ₂	1556	0.015	0.18	0.73	1.4	2.0	---
Cl ₂	557	0.075	0.37	1.00	---	---	---

In this calculation $\omega\tau$ was set equal to 100 and $\frac{d}{dT}(\ln K^{10})$ equal to 0.01. For this range of $\omega\tau$ the amplification per wavelength is approximately proportional to $(\omega\tau)^{-1}$ or the amplification per unit length is independent of frequency.

The equations derived above also apply to a mixture of a diatomic gas with a monoatomic gas such as N₂-He. In pure N₂ the relaxation time is of the order of atmosphere seconds. To observe appreciable gain, measurements would have to be made at such low frequencies and long wavelengths that it would be very difficult to conduct the needed measurements in the laboratory. By adding He or H₂ molecules, the frequencies where appreciable gain is expected are more accessible.

When the sound wave is propagated through a metastable state with more than one excited vibrational mode the problem becomes more complicated. However, the change in \dot{E}_a due to the sound wave perturbation can be obtained by differentiating Equation 5. The frequency dependent specific heat similar to Equation 14 is given by

$$C_v(\omega) = C_\infty + C_a \frac{\Delta T_a}{\Delta T} + C_b \frac{\Delta T_b}{\Delta T} + \dots, \quad (17)$$

where

$$C_i = R \frac{\left(\frac{\theta_i}{T_i}\right)^2 e^{-\theta_i/T_i}}{(1 - e^{-\theta_i/T_i})^2} \quad (18)$$

$i=a,b,\dots$. Using Equation 17 and a similar equation for $C_p(\omega)$, Shields has calculated the sound amplification in CD_4 and a mixture of N_2-CO_2-He . The results are shown in Table 2 and Figure 1 of Reference 10.

EXPERIMENTAL CONDITION FOR OBSERVING THE AMPLIFICATION

As discussed in the preceding section, an electrical discharge in low pressure N_2 will result in overpopulation of excited vibrational states. The electric circuit used in this experiment to produce the discharge is able to deposit approximately 2.5 joules of energy in the gas. Assuming about 70% of this goes to vibrational energy,²⁸ the vibrational temperature was estimated to be of the order of a few thousand degrees Kelvin depending on the volume and pressure of the gas.

After the discharge has been turned off, according to Margottin Maclou, Dovennette, and Henry²⁸, the vibrationally excited gas will relax in three different ways. First is relaxation due to collisional

de-excitation in which the vibrational energy is transferred into translational energy. Second is relaxation due to wall collision in which the vibrational energy is transferred to the wall. Third is relaxation due to spontaneous radiation. Since N_2 has no dipole moment, the latter process is not involved in its relaxation.

Let τ_v and τ_{vc} be the relaxation times for transferring vibrational energy to translation and to the tube wall, and let τ_{tc} be the relaxation time equating the gas translational temperature with the wall temperature by thermal conduction. The variation of the translational temperature with time then depends upon τ_v , τ_{vc} , and τ_{tc} . Margottin-Maclou, et al.²⁶ found that in carbon monoxide $\tau_{vc} = \tau_{tc}$. We will assume that the two are approximately equal in N_2 also.

For the experiment proposed here, it will be necessary for the sound period, P , to be less than either τ_v , τ_{vc} , or τ_{tc} . Unless P is less than τ_{tc} the propagation will be isothermal and no relaxation effects will be observed. If P is not less than τ_v and τ_{vc} the metastable state will not persist over a number of sound vibrations. However, if P/τ_v is small $\omega\tau_v$ will be large and the gain per wavelength will be small (Notice the gain per wavelength is inversely proportional to $\omega\tau_v^{10}$). In addition, the total path length for the sound in the gas, and thus the total gain, is restricted by the condition that the transit time for the sound wave through the gas must be small compared to the vibrational relaxation time, τ_v , and/or τ_{vc} . Otherwise the overpopulation of excited vibrational states decays away before the sound wave can traverse the gas.

A final constraint must be placed upon the ratio τ_v/τ_{tc} . Unless this ratio is considerably more than one, hot spots are likely to develop in the gas and there is not time for spatial variations in temperature to be smoothed out by conduction before the vibrational excitation dies away. What the above means is that there is a very limited range of values of τ_v/τ_{tc} for which it will be possible to observe the expected gain.

Table 2 shows the value of τ_v and τ_{tc} (in milliseconds) for gases used in this study. Values of τ_v were taken from the work of Frev, et al.¹⁶ and τ_{tc} 's were calculated using the formula^{26,27}

$$\tau_{tc} = \frac{1}{\xi^2 K}, \quad (19)$$

where $K = \frac{k}{\rho C_v}$ is the coefficient of diffusivity ($\text{cm}^2 \text{s}^{-1}$), and $(a\xi)$ is the first zero of the Bessel function $J_0(a\xi)$ where a is the tube radius. In this calculation, the thermal conductivity, k , for pure nitrogen was taken from the Table of Thermal Properties of Gases, National Bureau of Standards, Circular 564, Nov. 1955, and for mixtures were calculated using equation 8.2-40 of Hirschfelder, Curtis, and Bird²³. τ_v and τ_{tc} were calculated at pressure 40 Torr in a 0.615 cm radius tube.

TABLE 2

Vibrational relaxation time τ_v and translational relaxation time τ_{tc} (both in milliseconds) for several gases, calculated at pressure of 40 Torr in a tube of 0.615 cm in radius.

Gas	τ_v (ms)	τ_{tc} (ms)
Pure N_2	19×10^3	11.56
25He-75 N_2	520	7.16
50He-50 N_2	260	3.79
10 H_2 -90 N_2	52.8	11.16
20 H_2 -80 N_2	26.4	8.78

As we can see from Equation 19, τ_{tc} is directly proportional to pressure and inversely proportional to the squared tube radius. Since τ_v is inversely proportional to pressure, the desired ratio of τ_v/τ_{tc} can be obtained by choosing the proper tube radius and gas pressure. However, there is a high pressure limit due to the difficulty in getting a uniform electrical discharge in a gas above a few tens of Torr pressure. Increasing the pressure also increases the energy needed in the discharge to produce the high vibrational temperature.

CHAPTER III

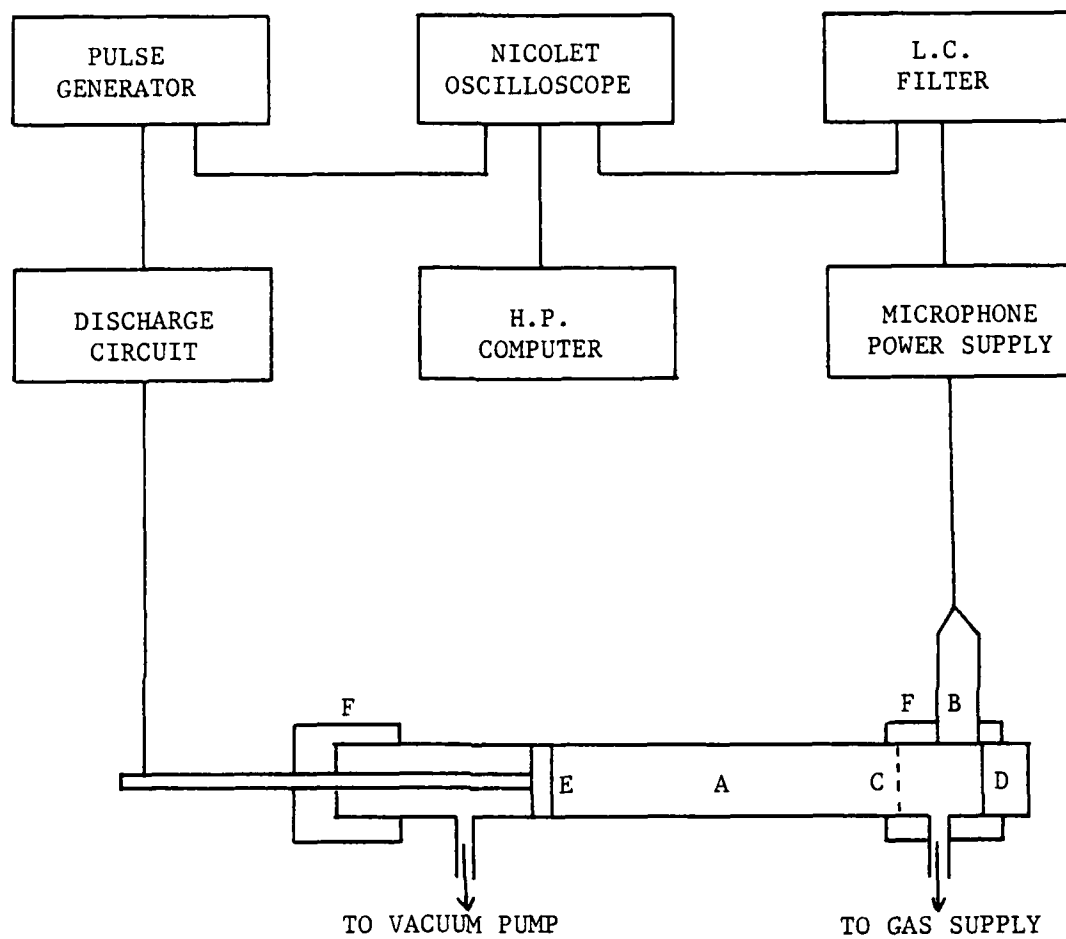
EXPERIMENTAL

APPARATUS

The apparatus used in making the sound absorption and velocity measurements is diagrammed in Figure 1. The different components are described below.

Sound Tube

The sound tube, which contained a movable reflector, was a pyrex tube, about 100 cm long. Two different size tubes, 2.54 cm and 1.23 cm in inner diameter, were used. One end of the glass tube was joined coaxially to a short section of a brass tube. A receiving microphone was mounted on the side and a stationary reflector was mounted in the end of the brass section. (In experiments with stable gases the stationary reflector was replaced by a speaker which served also as a reflector.) The brass section was connected to the gas handling system by means of 0.25 inch in diameter copper tubing. At the junction between the glass tube and the brass section a metal screen made of a piece of copper wire mesh of about 25 cm² was mounted terminating the electrical discharge. The brass section together with the gas handling system were grounded. The other end of the tube was terminated with the second brass section, the end of which contained an O-ring seal through which the rod moving the reflector passed. This second brass section was connected to the gas



- A. GAS SAMPLE
- B. MICROPHONE AND AMPLIFIER
- C. SCREEN
- D. STATIONARY REFLECTOR/SPEAKER
- E. MOVABLE REFLECTOR
- F. BRASS HOLDER

FIGURE 1. DIAGRAM OF MEASURING SYSTEM

handling system by means of a 3/8 inch pyrex tube, and the reflector was connected to the positive terminal of the discharge circuit. This end of the sound tube was electrically isolated from the grounded end by means of plastic rod supports. The movable reflector and the screen served as a pair of electrodes between which the discharge occurred. The gas in the region between the electrodes was rapidly heated by the discharge whereas the gas in the region between the screen and the stationary reflector remained cool. The length of this cool section was adjusted producing the optimum sound pulse.

Microphone and Speaker

The microphone was mounted on the side of the grounded brass section, approximately 3.5 cm away from the screen, and immediately in front of the stationary reflector. Its face was parallel to the tube axis and was adjusted so that the microphone surface protruded only slightly into the sound tube avoiding sound reflections. In experiments with the 2.54 cm tube, the microphone was an adapted one inch Bruel-Kiaer (B-K) condenser microphone. The microphone was connected to the preamp through a vacuum feed-through that was sealed through the wall of the brass tube with a Swagelock fitting. In experiments with the 1.23 cm tube, the microphone was an adapted one-half inch B-K condenser microphone with diaphragm made of Mylar of 1.27×10^{-3} cm thick. Both microphones were powered by a B-K type 2801 power supply and their outputs were preamplified by a B-K type 2615 cathode follower amplifier.

The speaker used in experiments on stable gases was one previously used by Bass and Detsch in their investigation on the sound amplification in gas phase reaction systems.¹¹ The polarizing voltage of the speaker was maintained by a Heathkit model IP-17 regulated power supply and driven by tone bursts generated by a GR model 1396B tone burst generator and amplified by a Hewlett Packard (HP) Model 467A amplifier.

Discharge Circuit

The discharge circuit used in this experiment was a modification of the SCR discharge circuit designed by M.M.T. Loy and P.A. Roland.²⁸ The circuit diagram is shown in Figure 2. The original 16 F Mylar capacitor was replaced by three 10 μ F Micamold capacitors set in parallel. The pulse transformer was an automobile ignition coil, Delco D523, and the SCR a type 50 RIA100. The 2 volt-1 μ second trigger pulse was generated by a HP model 214A pulse generator. The transformer was placed near the

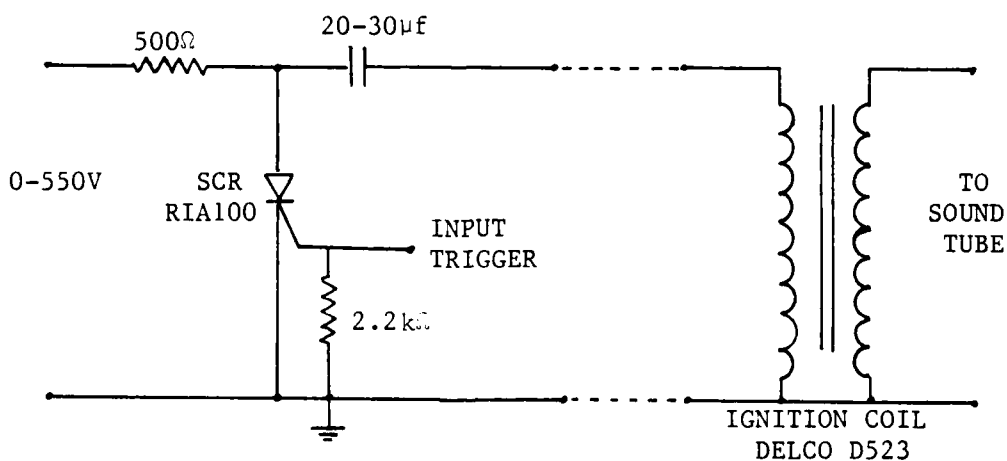


FIGURE 2. DISCHARGE CIRCUIT

sound tube and connected to the SCR circuit, housed in a separate box, by a shielded cable. The discharge circuit was able to produce a discharge of approximately 2.5 joules in 1.23 cm tube, 30 to 50 cm long at pressure of 40 Torr.

Electronics

An externally-triggered model 214A HP Pulse generator was used to trigger the discharge and the Nicolet oscilloscope trace. The large capacitor, at relatively high voltage, then discharged through the primary of the transformer. The voltage pulse from the secondary of the transformer produced the discharge in the gas sample and a metastable state with an overpopulation of excited vibrational states resulted.

Efforts to generate the sound pulse in the metastable gas using a speaker in the end of the discharge tube were unsuccessful due to the masking effects of the discharge on the sound wave and damage done to the transducers by the discharge. These difficulties were finally solved by placing a metal screen a few cm from the speaker/reflector, as discussed in the preceding section. In this configuration, the sound pulse was generated by a sudden increase in temperature of the gas in the region where the discharge occurred. The increase in temperature caused a pressure increase which expanded into the cool section. The expansion produced a pulse which reflected back and forth in the tube. The pressure pulse was detected each time it was reflected from the microphone end of the tube. The signal from the microphone, after being amplified, was passed through an Ithaco 4302, 24 dB/octave filter and then applied to the vertical input of the Nicolet digital oscilloscope.

The signal thus displayed on the oscilloscope screen was stored in the scope memory and finally transferred to a HP model 85 computer to be analyzed.

The sampling time of the oscilloscope may be adjusted from 500 nsec to 200 sec per point. In this experiment the sampling time was set at 10 usec per point. The total number of points recorded and stored by the oscilloscope was 4096 points. The data points of reflected pulses were transferred to the computer and Fourier analyzed one pulse at a time.

Gas Handling System

The system was evacuated with a Welch Duo seal fore pump which was connected to the manifold of the gas handling system. The pressure of the system was measured with M.K.S. Baratron type 170 M-25 and PDR-D-1 transducers. The first being used for the pressure range up to 10 Torr and the latter used from 10 to 100 Torr. The fore pump was capable of evacuating the system to less than 2 millitorr. Gas samples were stored in storage tanks and connected to the sound tube through a manifold and a series of valves.

EXPERIMENTAL PROCEDURE

The absorption and velocity measurements were made in pure N_2 and mixtures of N_2 with He and H_2 . The gas mixtures were made using the method of partial pressure. A storage tank was alternately filled with N_2 and He/ H_2 until the desired concentration at total pressure of one atmosphere was established. The gases were then allowed to mix at least one day before measurement was made. Since the relaxation time and the

maximum relaxation frequency of N_2 are very sensitive to the concentration of impurities, especially water vapor, it was necessary to ensure the purity of the gas under consideration. For this purpose the following steps were followed. Prior to making any measurement, the system was thoroughly evacuated and the combined outgassing and leaking rate was frequently checked. (This system was able to be evacuated to about 2 milli Torr and the combined outgassing and leaking rate was less than 0.5 milli Torr per minute at a pressure of 20 Torr.) The pressure in all pipes connecting the storage tanks and the manifold of the gas handling system was always kept lower than the pressure of interest in the sound tube to avoid impurity leaks into the sound tube. The sound tube was then flushed several times with the gas sample. In doing so, the evacuated sound tube was filled with the test gas to about 100 Torr in pressure and re-evacuated. This was repeated several times. The electronic equipment, except the discharge circuit, was warmed up before measurements were made. The band-pass filter was set to pass frequencies from 2 to 6.3 kHz.

When everything was ready, the pump was closed off from the system and the gas sample to be tested was introduced into the tube until the desired pressure was obtained. After thermal equilibrium with the tube wall was established, the pressure and the tube wall temperature were recorded and the discharge circuit was turned on with the capacitor voltage set at a desired value. To avoid impurities generated by the sparks, the sound signal was taken from the first spark that gave the

expected signal shape. To further ensure the purity of the gas sample under consideration, every sound signal recorded was for a fresh gas sample that had been in the tube no longer than 60 seconds. The procedure discussed above was repeated for measurements at different pressure and tube lengths (the distance between reflectors). For a given set of these parameters, data was taken for approximately nine discharges and averaged.

FOURIER TRANSFORM

The absorption and velocity of sound at a given frequency were determined by Fourier analyzing the sound pulse after each reflection. For this purpose, the signal detected by the microphone was filtered and then digitized by the oscilloscope. The starting time and ending time for each of the reflected pulses were read visually off the oscilloscope and stored in the computer. The digitized signal between these two times was then transferred to the computer and Fourier analyzed. The fundamental frequency in the analysis was set at 390.6 Hz by setting the time interval covered in each analysis at 256 points or 2.56×10^{-3} seconds. If, for example, there were N digitized points between the beginning and end of the pulse as read visually from the oscilloscope, the signal Fourier analyzed would consist of N points transferred from the oscilloscope memory followed by 256-N zero points. The sampling period, $T = 10$ μ sec, easily satisfied the condition that $1/T = 2 \times$ maximum frequency since the highest frequency with an appreciable component in the pulse was 6 kHz.^{29,30} The number of points, 256, was limited by the time

required by the computer to make the analysis. The 390 Hz frequency spacing, however, was quite adequate for the purpose of this experiment.

The average sound velocity for one round trip in the tube was obtained by the equation

$$V_n = \frac{2\pi n(2L)}{N\Delta t\Delta\phi}, \quad n = 0, 1, 2, \dots \quad (20)$$

where N is the number of sample points (256), Δt is the sampling period (10 μ sec), $\Delta\phi$ is the change in phase after the pulse had travelled one round trip in the tube, L is the distance between the microphone and reflector. $n/N\Delta t$ corresponds to the n th frequency component.

The amplitude of various frequency components in successive pulses are related by the equation

$$A_2 = A_1 e^{-(2L\alpha + \beta)} \quad (21)$$

where α is the absorption coefficient (neper/cm) and β is the effective reflection coefficient. By making measurements at more than one value of L , both α and β can be determined. Once the reflection coefficient is determined, α can be determined from the amplitude without varying the length of the sound path.

A computer program developed by the Hewlett Packard Company³¹ was modified in order to determine the sound absorption and velocity of each frequency component of interest directly from the amplitude and phase obtained from the Fourier analysis. The computer program was developed by using the Fast Fourier Transform (FFT) algorithm that transformed 2N

sample points by means of an N point transform. The results of this procedure were checked by direct measurements of the sound absorption and velocity of the pulse after it passed through the filter set at a single frequency. The two methods gave results that agreed within experimental error. The computer program used to Fourier analyze the sound signal is presented in the appendix.

CHAPTER IV

EXPERIMENTAL RESULTS

RESULTS OF MEASUREMENTS IN STABLE GASES

Sound amplification and translational temperature were obtained by comparing the measured absorption and velocity in excited gases with the absorption and velocity calculated for the unexcited gases using the numerical solution discussed above. To check the accuracy of these calculations the sound absorption and velocity were measured in unexcited nitrogen gas and mixtures of 25% He - 75% N₂, 50% He - 50% N₂ of various pressures and frequencies. In these measurements, the gas was contained in a cylindrical glass tube 2.54 cm inside diameter. A tone burst, produced by the speaker in the end of the tube was allowed to propagate back and forth between the speaker and the reflector while the amplitude was monitored by the microphone. The amplitude of the tone burst after it was reflected m times is given by

$$A = A_0 e^{-(2L\alpha + \beta)m} \quad (22)$$

where α , β and L have been defined previously. The absorption, α , and the coefficient of reflection, β , were obtained by plotting on a log scale the amplitude, A versus L , with m as a parameter. A sample of these plots is shown in Figure 3. The straight line plots of amplitude versus distance for the various value of m all have the same slope

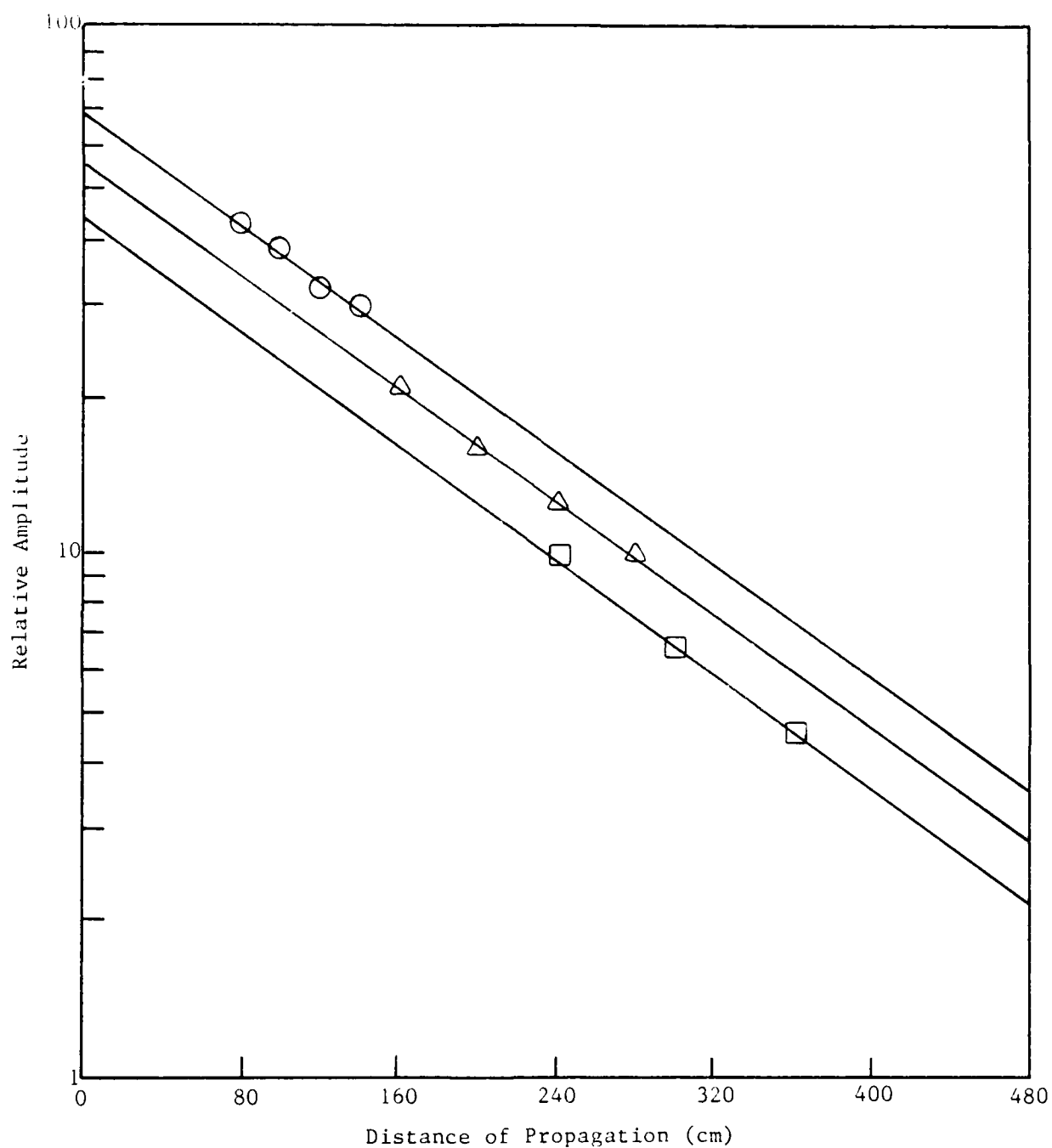


FIGURE 3. Sound amplitude vs distance of propagation in stable N_2 at pressure = 20 Torr; frequency = 2 kHz; tube radius = 1.27 cm. \circ = $m=1$ (first reflection); \triangle = $m=2$ (second reflection); \square = $m=3$ (third reflection).

corresponding to α , and they are displaced from each other by an amount corresponding to β . The results obtained from a least square fit of a straight line to the data are given in Tables 3 and 4 along with the calculated values. From comparison of values in these tables it is concluded that individual measured and calculated values of absorption differ by at most $\pm 2\%$. Note that relaxation absorption in nitrogen gas at room temperature is very small. This factor was not included in this calculation. The coefficient of reflection, β , in N_2 , 25% He - 75% H_2 , and 50% He - 50% N_2 , averaging overall pressures and frequencies of interest, were found to be 0.18 ± 0.03 , 0.18 ± 0.02 , and 0.15 ± 0.02 respectively.

The velocity was obtained by measuring the time the sound took to travel a distance of $2L$ (one round trip) in the tube. The results are compared with theoretical in Tables 5 and 6. From comparison of these values it is concluded that the maximum difference in individual measured and calculated values of velocities is $\pm 0.3\%$.

SOUND ABSORPTION AND VELOCITY IN EXCITED METASTABLE STATE GASES

Absorption and velocity measurements in excited metastable state gases were made with two different tube sizes, 2.54 cm and 1.23 cm inside diameter. Gases used with the bigger tube were pure N_2 and mixtures of 25% He - 75% N_2 and 50% He - 50% N_2 all at pressures of 20 Torr. In experiments with the smaller tube, the above gases and a 10% H_2 - 90% N_2 mixture at pressures of 20 and 40 Torr were used. Experiments were begun by discharging the capacitor through the primary of the induction coil,

TABLE 3
 Sound absorption (dB/cm) in stable gases at various pressures.
 Frequency = 2 kHz, tube radius = 1.27 cm.

GAS	P = 20 Torr		P = 30 Torr		P = 40 Torr		P = 60 Torr	
	obs.	calc.	obs.	calc.	obs.	calc.	obs.	calc.
N ₂	.0564	.0562	.0457	.0454	.0385	0.392	.0308	.0319
25He-75N ₂	.0575	.0586	.0494	.0476	.0411	0.411	-----	-----
50He-50N ₂	.0620	.0616	.0498	.0501	.0435	0.432	.0351	.0352

TABLE 4
Sound absorption (dB/cm) in stable gases at various frequencies.

Pressure = 20 torr, tube radius = 1.27 cm.

GAS	f = 1.6 kHz		f = 2 kHz		f = 3.15 kHz		f = 4 kHz	
	obs.	calc.	obs.	calc.	obs.	calc.	obs.	calc.
N ₂	.0509	.0502	.0564	.0562	.0706	.0707	.0796	.0796
25He-75N ₂	-----	-----	.0575	.0585	.0727	.0735	-----	-----
50He-50N ₂	.0553	.0552	.0620	.0612	.0767	.0772	.0880	.0871

TABLE 5

Sound velocity in stable N_2 at various frequencies.

Pressure = 20 Torr, tube radius = 1.27 cm.

$f(\text{kHz})$	$V_{\text{obs.}} (\text{m/s})$	$V_{\text{calc.}} (\text{m/s})$
1.6	-----	342.4
2.0	342.8	343.1
3.15	345.2	344.3
4.0	344.8	344.8

TABLE 6

Sound velocity in stable 50% He - 50% N_2 mixture at various pressures.

$f = 2 \text{ kHz}$, tube radius = 1.27 cm.

$P(\text{Torr})$	$V_{\text{obs.}} (\text{m/s})$	$V_{\text{calc.}} (\text{m/s})$
20	468.5	469.7
30	471.6	471.8
40	472.4	473.1

thus producing the discharge in the gas sample. The electrical discharge rapidly increased the gas pressure in the region between the two electrodes (the region between the reflector and the screen; see Figure 1). The rapid increase in the pressure in the discharge region creates a pressure pulse at the hot-cold junction where the discharge terminated in the gas. This pulse travelled back and forth in the tube and was monitored by the microphone. The microphone response to the sudden pressure increase is illustrated in Figure 4. In this figure, positive pressure corresponds to a negative voltage. The spikes represent the sound pulse that reflected back and forth in the tube. The shape of the response curve without the superimposed pulses is the response of the microphone to the rapid increase in pressure that decays with a decay time equal to that for cooling the gas in the tube. By filtering and Fourier analyzing the signal, the amplitude and the phase of the different frequency components in the pulse as a function of time can be determined. Examples of a filtered signal are shown in Figures 5 and 6. The legend for these and other figures specifies the energy in the discharge in joule/mole. The energy was obtained by dividing the energy stored in the capacitor before discharge by the moles of the gas heated by the discharge. It is estimated that approximately 70% of this energy actually went into heating the vibrational modes of the gas.

The decreasing peaks in Figure 5 represent the sound pulse detected after it travelled successive round trips down the tube (a distance of 2 l). The first peak is the original pulse detected after it travelled a distance of approximately 4 cm through the cold region. The second peak

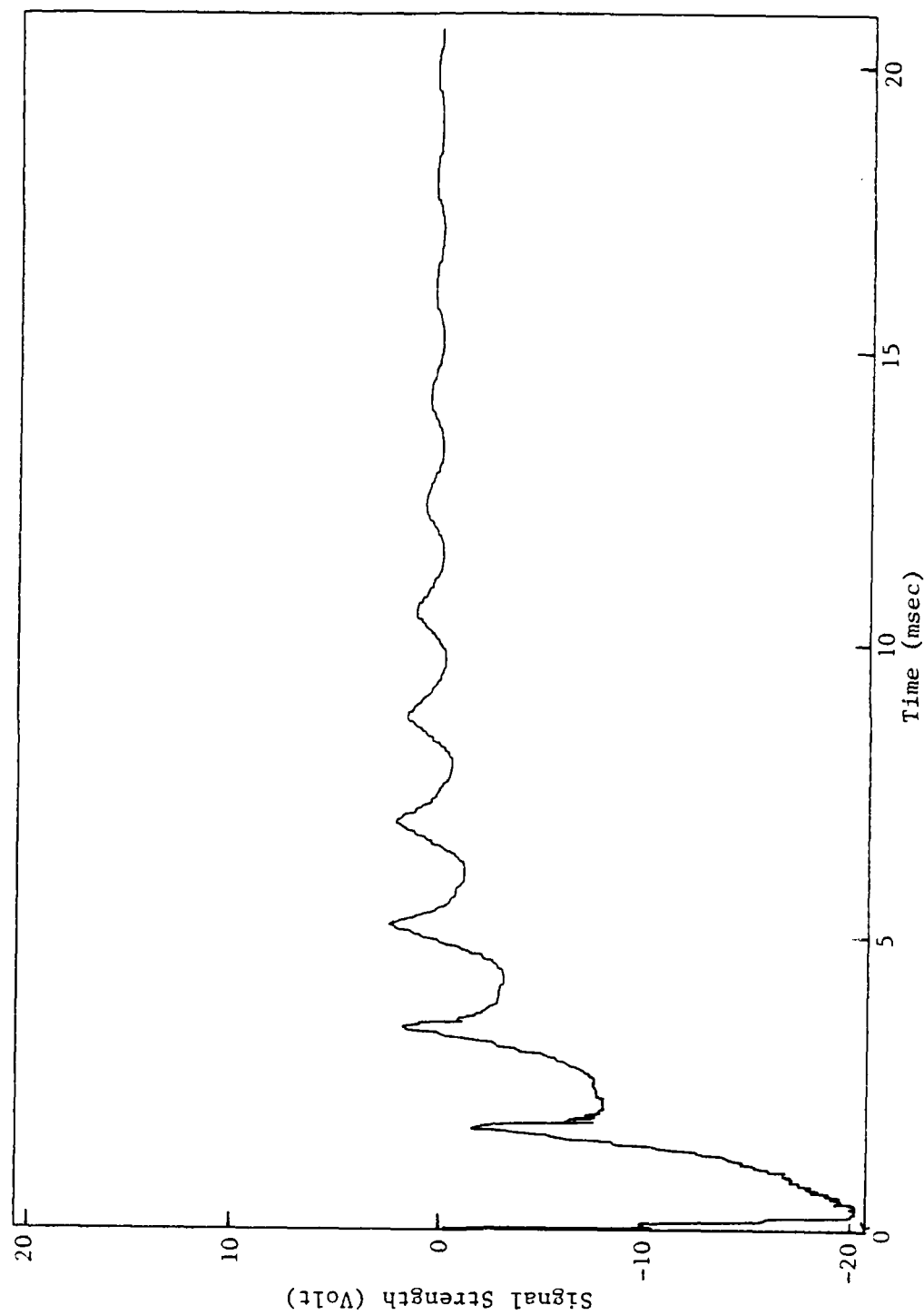


FIGURE 4. Unfiltered sound signal taken by Nicolet Oscilloscope set at 10 μ sec per point in pure N_2 at pressure = 40 Torr; tube radius = 0.615 cm; tube length = 30 cm; discharge energy = 49.3×10^3 joule/mole.

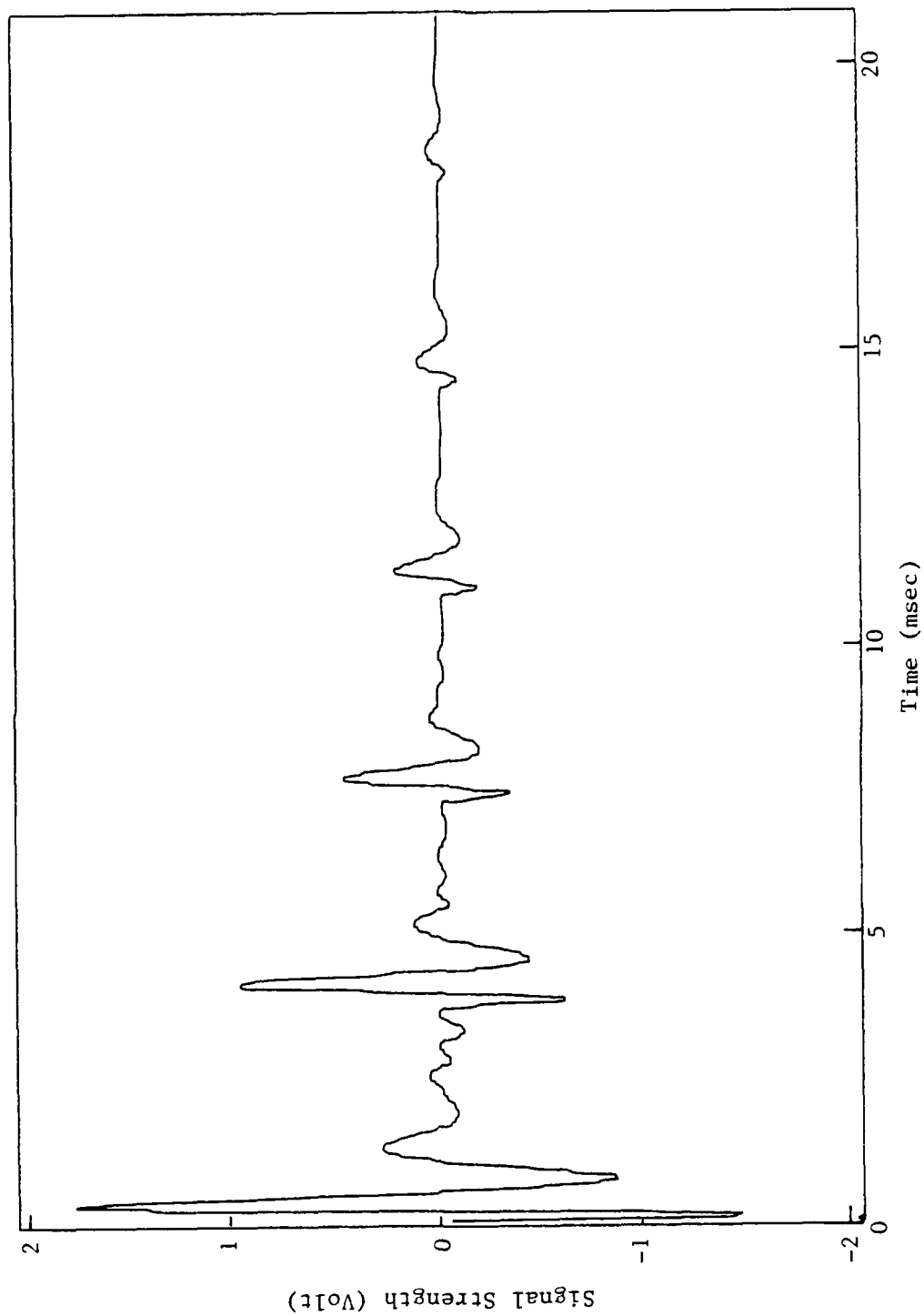


FIGURE 5. Sound pulse taken by Nicolet Oscilloscope set at 5 μ sec per point in pure N_2 at pressure = 40 Torr; tube radius = 0.615 cm; tube length = 30 cm; discharge energy = 49.3×10^3 joule/mole.

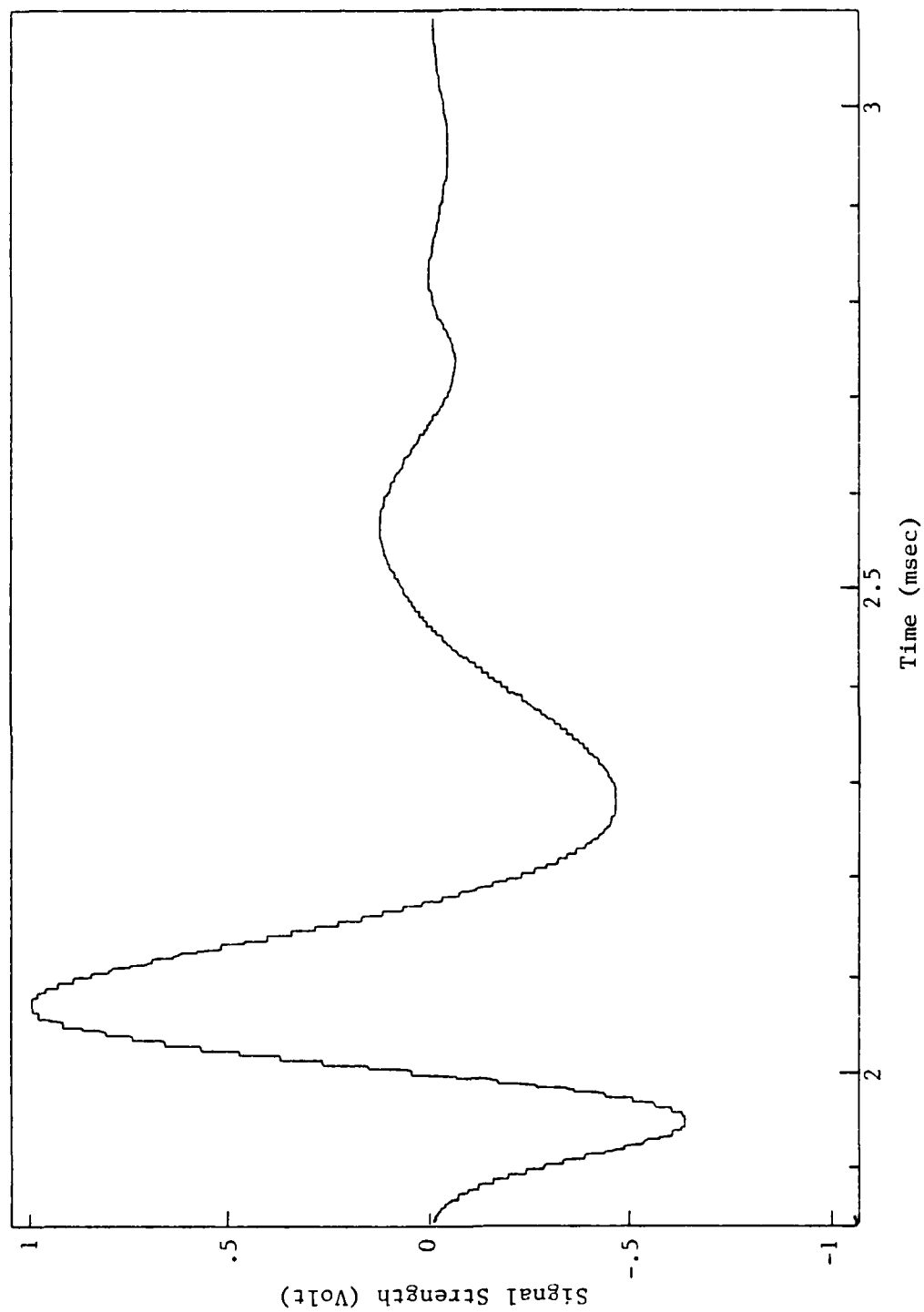


FIGURE 6. An expand display of a single one of the pulses in Figure 5, taken from 1.84 msec to 3.10 msec.

is the pulse detected after it travelled one round trip. The third peak is the pulse detected after it travelled two round trips, etc. The distance between two adjacent peaks corresponds to the time the pulse took to travel a distance of $2L$. In general the first pulse considered in our analysis was the one reaching the microphone after travelling one round trip in the tube.

The average absorption per unit length between any two successive reflections is given by

$$\alpha = (-\ln \frac{A_{m+1}}{A_m} - \beta)/2L; m = 1, 2, 3, \dots \quad (23)$$

The absorption determined by Equation 23 was assumed to be the absorption at a time equal to

$$t = (t_m + t_{m+1})/2, \quad (24)$$

where t_m is the time after the pulse was reflected m times, taken at the center of the pulse. In the case that the sound pulse is amplified, α is time dependent because the vibrational temperature is time dependent. In experiments with the 2.54 cm diameter tube, β was obtained from the experiment on stable gases as discussed in the preceding section. With a 1.23 cm diameter tube, attempts to determine β by using a tone burst from the speaker failed because the sound attenuation at low pressure was too large. The tone burst decayed out very rapidly. In this case, a sound pulse generated by the discharge was used instead of the tone burst. β was determined from the intercept of the plot of the logarithm of amplitude

versus distance, L , as discussed previously. In this case, of course, the discharge circuit was adjusted so that the energy per unit volume in the discharge remained constant for all reflector separations. Discharges were chosen for different reflector distances, L , so that the amplitudes for the first reflected pulse were the same. Table 7 shows the coefficient of reflection in Neper/reflection averaged over frequency components from 1.9 to 4.2 kHz, observed in gases used in this experiment.

TABLE 7

Coefficient of reflection (β) observed in experiments with 1.23 cm diameter tube.

GAS	(Neper/reflection)	
	P = 20 Torr	P = 40 Torr
N_2	0.16 ± 0.07	0.15 ± 0.03
25He-75 N_2	0.05 ± 0.03	0.09 ± 0.02
50He-50 N_2	0.10 ± 0.04	0.11 ± 0.04
10 H_2 -90 N_2	0.03 ± 0.01	0.07 ± 0.01

Results of Measurements in Excited Metastable Pure Nitrogen

Sound absorption and velocity observed in excited metastable pure nitrogen are presented in Tables 8-13. Table 8 gives the average absorption coefficient over the total time the pulse was observed. Also

TABLE 8

Time average sound absorption ($\Delta\alpha$) in pure excited metastable N_2 . Values are given in dB/cm. $\alpha_{calc.}$ (in this table and the following tables) were obtained from Shields' numerical solution assuming the temperature of the gas was that obtained from the velocity measurements (301°K). $\Delta\alpha = \alpha_{calc.} - \alpha_{obs.}$

Frequency (Hz)	Tube radius = 1.27cm				Tube radius = 0.615cm							
	P = 20 Torr				P = 20 Torr				P = 40 Torr			
	$\alpha_{obs.}$	$\alpha_{calc.}$	$\Delta\alpha$		$\alpha_{obs.}$	$\alpha_{calc.}$	$\Delta\alpha$		$\alpha_{obs.}$	$\alpha_{calc.}$	$\Delta\alpha$	
1563	0.049	0.050	0.001		0.104	0.105	0.001		-----	0.073	-----	
1953	0.052	0.056	0.004		0.114	0.117	0.003		0.075	0.083	0.008	
2344	0.061	0.061	-----		0.123	0.128	0.005		0.086	0.089	0.003	
2734	0.068	0.066	-0.002		0.139	0.138	-0.001		0.091	0.096	0.005	
3125	-----	-----	-----		0.141	0.148	0.007		0.095	0.103	0.008	
3515	-----	-----	-----		0.154	0.156	0.002		0.101	0.109	0.008	
3906	-----	-----	-----		0.156	0.165	0.009		0.111	0.115	0.004	
4297	-----	-----	-----		0.174	0.174	-0.001		0.120	0.121	0.001	

TABLE 9

Sound absorption (α) as a function of time in pure excited metastable N_2 .
 pressure = 40 Torr; tube radius = 0.615 cm; discharge energy = 22.4×10^3 joule/mole.

Values are given in dB/cm. $\Delta\alpha = \alpha_{\text{calc.}} - \alpha_{\text{obs.}}$

Time (ms) f (Hz)	3.91		6.23		8.72		$\alpha_{\text{calc.}}$ at T = 301°K
	$\alpha_{\text{obs.}}$	$\Delta\alpha$	$\alpha_{\text{obs.}}$	$\Delta\alpha$	$\alpha_{\text{obs.}}$	$\Delta\alpha$	
1953	0.075	0.008	0.075	0.008	0.075	0.008	0.083
2344	0.087	0.002	0.086	0.003	0.086	0.003	0.089
2734	0.090	0.006	0.091	0.005	0.091	0.005	0.096
3125	0.097	0.006	0.095	0.008	0.095	0.008	0.103
3515	0.101	0.008	0.101	0.008	0.100	0.009	0.109
3906	0.111	0.004	0.111	0.004	0.111	0.004	0.115
4297	0.120	0.001	0.120	0.001	0.120	0.001	0.121

TABLE 10

Sound absorption (α) as a function of time in excited metastable state N_2 at

$P = 20$ Torr; tube radius = 1.27 cm; discharge energy = 10.6×10^3

joule/mole. $\Delta\alpha = \alpha_{\text{calc.}} - \alpha_{\text{obs.}}$ Values are given in dB/cm.

time (ms) f (Hz)	4.77		7.61		10.45		13.31		calc. at $T=304^\circ\text{K}$
	$\alpha_{\text{obs.}}$	$\Delta\alpha$	$\alpha_{\text{obs.}}$	$\Delta\alpha$	$\alpha_{\text{obs.}}$	$\Delta\alpha$	$\alpha_{\text{obs.}}$	$\Delta\alpha$	
1563	0.050	0.0	0.048	0.002	0.048	0.002	0.050	0.0	0.050
1953	0.054	0.002	0.051	0.005	0.050	0.006	0.055	0.001	0.056
2344	0.062	-0.001	0.062	-0.001	0.061	0.0	0.062	-0.001	0.061
2734	0.069	-0.003	0.071	-0.005	0.066	0.0	0.070	-0.004	0.066

TABLE 11

Sound velocity in m/s as a function of time in excited
 metastable N_2 at Pressure = 20 Torr; tube length = 40 cm;
 tube radius = 1.27 cm; discharge energy = 10.5×10^3 joule/mole.

time (ms) f (Hz)	4.77	7.61	10.45	13.31	V_{calc} at $T=304.5^\circ K$
1562	350.7	350.5	350.2	350.3	348.7
1953	350.8	350.4	350.4	350.6	349.5
2344	350.5	350.2	350.2	350.6	350.0
2734	349.7	349.5	350.1	350.0	350.4

TABLE 12

Sound velocity in m/s as a function of time in excited
 metastable N_2 at Pressure = 20 Torr; tube length = 40 cm;
 tube radius = 0.615 cm; discharge energy = 33.4×10^3 joule/mole.

time (ms) f (Hz)	3.87	6.20	8.47	v_{calc} (ms) at $T=304^\circ K$
1953	342.6	342.2	343.0	342.8
2344	342.9	343.1	344.0	343.9
2734	343.9	344.1	344.9	344.8
3125	344.9	345.1	346.1	345.4
3515	345.5	345.8	346.8	346.0
3906	345.8	346.0	347.2	346.5
4297	346.1	346.5	347.7	346.9

TABLE 13

Sound velocity in m/s as a function of time in excited
metastable N_2 at Pressure \approx 40 Torr; tube length = 40 cm;
tube radius = 0.615 cm; discharge energy = 16.7×10^3 joule/mole.

time (ms) f (Hz)	3.91	6.23	8.72	v_{calc} (ms) at $T=301^\circ K$
1953	344.6	344.3	345.0	344.7
2344	344.9	344.8	345.1	345.5
2734	345.2	345.4	345.5	346.0
3125	345.7	345.9	346.1	346.6
3515	346.1	346.2	347.5	346.9
3906	346.4	346.3	347.6	347.3
4297	346.6	346.7	347.1	347.6

given in Table 8 are values of the absorption calculated for stable nitrogen at the equilibrium pressure and the temperature determined from the velocity measurements. Measured values of the absorption and velocity in Tables 8-13 are the average of nine or more individual measurements. From the standard deviation in these individual measurements it is estimated that the measured values given in Table 8 are good to $\pm 3\%$. $\Delta\alpha$ is the difference between the observed and calculated absorption ($\Delta\alpha = \alpha_{\text{calc.}} - \alpha_{\text{obs.}}$).

The gain ($\Delta\alpha$) in dB/cm should be frequency independent.¹⁰ Making use of this fact, by averaging $\Delta\alpha$ over all frequency components in Table 8, we found $\Delta\alpha$ of approximately 0.003 dB/cm in the bigger tube, and 0.004 dB/cm and 0.005 dB/cm in the smaller tube at pressures of 20 and 40 Torr respectively. This difference is only slightly more than experimental error. If it is due to relaxation, it should be changing with time. Therefore, in Tables 9-13 the absorption and velocity between individual reflections is tabulated as a function of the time at the midpoint of the interval considered. Any change in velocity and absorption with time was within experimental error.

At first sight it might be somewhat surprising that the temperature of the nitrogen gas was not changed more by the discharge. (The velocity measurements show the gas temperature to be only a few degrees above the tube wall (room) temperature.) In later experiments with mixtures, the discharge was found to heat the gas from 10 to 40°C. However, a closer look at the properties of pure nitrogen explain the absence of the heating. Michan's calculations¹⁵ from measured electron cross sections

in nitrogen indicate that for the voltages and pressures of our discharge, practically all of the discharge energy goes into vibration. Further, the vibrational relaxation time in pure nitrogen is so much greater than the thermal diffusion time in the gas that the vibrational energy is conducted to the walls without appreciably heating the gas.

Though the difference between measured and calculated absorption is small, the observed values were consistently smaller than calculated as would be expected if amplification were present. If this difference between measured and calculated absorption is due to relaxation it is larger than Shields' calculation.¹⁰ Assuming the vibrational temperature is between 1000 and 2000°K during the measurement, the gain would indicate a relaxation time in nitrogen of the order of a few milliseconds per atmosphere. This relaxation time is considerably shorter than the relaxation time in pure nitrogen and, for the estimated amount of energy in the discharge, the vibrational temperature could not exceed 3000°K.

The small discrepancy is thus unexplained, and is assumed to be due to a consistent experimental error. In experiments on mixtures of nitrogen with helium and hydrogen, which will be discussed later, $\Delta\alpha$ here observed will be used as a correction factor.

The uncertainty in velocity at a particular time, estimated from the standard deviation in the individual measurements, is approximately $\pm 0.4\%$. In what follows, the temperature of the gas will be obtained by comparing measured velocities with values calculated as functions of

temperature by numerical solutions.²³ Temperatures obtained in this way are estimated to be accurate to $\pm 1^\circ\text{C}$.

Results of Measurements in He-N₂ Mixtures

Sound absorption measurements in He-N₂ mixtures are presented in Tables 14-17. The variation in $\Delta\alpha$ with time in 50% He - 50% N₂ mixtures is plotted in Figure 7.

The addition of He molecules to N₂ gas drastically reduces the relaxation time of N₂. In a 25% He - 75% N₂ mixture the relaxation time is reduced to about 1040 and 520 milliseconds at pressures of 20 and 40 Torr respectively, and in a 50% He - 50% N₂ mixture to 520 and 260 milliseconds.¹⁶ However, compared to the thermal conduction relaxation time (τ_{tc}) which is of the order of 10 milliseconds, the ratio of τ_v and τ_{tc} is still large. The vibrational energy transferred into translation during the transit time is still small, and the gain $\Delta\alpha$ is expected to be so small that it will be difficult to observe. The observed value of $\Delta\alpha$ given in Tables 14 and 15 was corrected by subtracting the $\Delta\alpha$ observed in pure nitrogen. When this is done, no appreciable gain is observed in the 25% He - 75% N₂ mixtures, nor is there an observable change in it with time.

However, in the 50% He - 50% N₂ mixtures there is a measurable gain. Averaged overall frequency components in Table 16, $\Delta\alpha$, after correcting by subtracting $\Delta\alpha$ observed in N₂, is 0.0018 dB/cm and 0.0074 dB/cm at pressures of 20 and 40 Torr respectively. The measured $\Delta\alpha$ values are

TABLE 14

Time average sound absorption (α) in excited metastable 25% He - 75% N₂ mixture.

Values are given in dB/cm.

Frequency (Hz)	Tube radius = 1.27cm		Tube radius = 0.615cm			
	P = 20 Torr		P = 20 Torr		P = 40 Torr	
	$\alpha_{\text{obs.}}$	$\alpha_{\text{calc.}}$	$\alpha_{\text{obs.}}$	$\alpha_{\text{calc.}}$	$\alpha_{\text{obs.}}$	$\alpha_{\text{calc.}}$
1563	0.048	0.052	0.117	0.115	0.078	0.077
1953	0.056	0.058	0.126	0.123	0.083	0.086
2344	0.064	0.064	0.136	0.134	0.091	0.094
2734	0.070	0.069	0.149	0.144	0.099	0.101
3125	-----	-----	0.155	0.154	0.109	0.108
3515	-----	-----	0.168	0.163	0.114	0.114
3906	-----	-----	0.172	0.172	0.117	0.121
4297	-----	-----	0.174	0.180	0.126	0.126

TABLE 15

Sound absorption (α) as a function of time in an excited metastable 25% He - 75% N₂

mixture. Pressure = 40 Torr; tube length = 30 cm; tube radius = 0.615 cm;

discharge energy = 16.7×10^3 joule/mole. Values are given in dB/cm.

Time (ms) f (Hz)	2.70	4.21	5.72	7.20	$\alpha_{\text{calc.}}$ at $T = 305^\circ\text{K}$
1953	0.085	0.082	0.082	0.083	0.086
2344	0.090	0.090	0.090	0.094	0.094
2734	0.096	0.100	0.103	0.109	0.101
3125	0.101	0.111	0.116	-----	0.108
3515	0.101	0.114	0.119	-----	0.114
3906	0.110	0.107	0.112	0.116	0.121
4297	0.120	0.113	0.114	0.128	0.126

TABLE 17

Sound absorption (α) as a function of time in excited metastable 50% He

- 50% N₂ mixture at pressure = 40 Torr; tube radius = 0.615 cm;

discharge energy = 33.5×10^3 . Values are given in dB/cm.

time (ms) f (Hz)	3.56		5.16		6.77		$\alpha_{calc.}$ at T=303°K
	$\alpha_{obs.}$	$\Delta\alpha$	$\alpha_{obs.}$	$\Delta\alpha$	$\alpha_{obs.}$	$\Delta\alpha$	
1953	.088	.002	.091	-----	.092	-----	.090
2344	.089	.010	.092	.007	.095	.004	.099
2734	.093	.014	.098	.009	.103	.004	.107
3125	-----	-----	-----	-----	-----	-----	-----
3515	.092	.028	.107	.013	.108	.012	.120
3906	.104	.023	.114	.010	.120	.007	.127
4297	.107	.026	.123	.007	.128	.005	.133

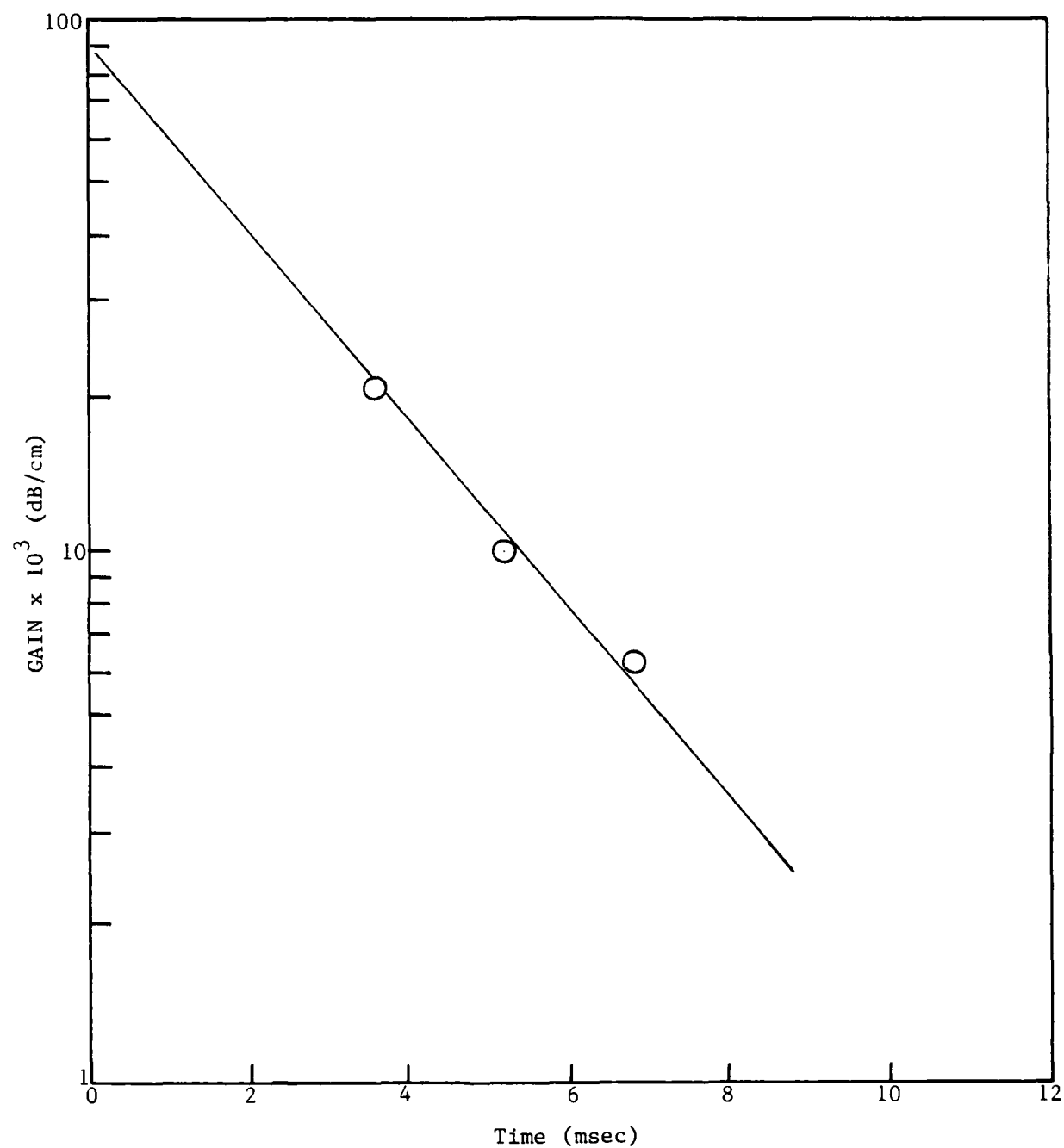


FIGURE 7. Gain ($\Delta\alpha$) as a function of time observed in 50% He - 50% N_2 mixture at pressure 40 Torr; tube radius = 0.615 cm; discharge energy = 33.4×10^5 joule/mole. Solid line is calculated gain setting $d(\ln \tau_V^{-1})/dT = 0.05$, $T_V = 6000^\circ K$ at 4 msec; $\tau_{tc} = 3.7$ msec.

plotted versus time in Figure 7. The magnitude and time dependence of this gain is compared to theory below.

Tables 18 and 19 give representative velocities measured in 25% He - 75% N₂ and 50% He - 50% N₂ mixtures at a pressure of 40 Torr and tube length of 30 and 40 cm respectively. This time the measured velocities show an appreciable increase in translational temperature. This indicates that, with the added He, a larger amount of the discharge energy is going directly into the translational energy of the gas mixture. The logarithm of the difference between the translational temperature (T_r) as determined from the velocity measurements and the tube wall temperature (T_w) is plotted versus time in Figures 8 and 9. The decay times calculated from the curves are compared with theoretical values of τ_{tc} in Table 20. The measured values of τ do have roughly the expected variation with pressure and tube radius as predicted by Equation 19.

In mixtures in which τ_{tc} was considerably longer than the time for the sound to make one round trip in the tube (25% He - 75% N₂ and 10% H₂ - 90% N₂ at a pressure of 40 Torr) the first data point of the translational temperature (see Figures 8 and 12) was far off the straight line. This was attributed to a non-uniform heating of the gas by the discharge. When τ_{tc} was long, compared to the transit time for the sound, there was not time for thermal diffusion to smooth out the hot and cold regions in the gas left by the non-uniform heating of the discharge. The hot spots evidently raised the measured sound velocity. At lower pressure the effect was not so obvious due both to the fact that the discharge was more uniform and the thermal diffusion time was shorter.

TABLE 18

Sound velocity in m/s as a function of time in excited metastable
 25% He - 75% N₂ mixture at Pressure = 40 Torr; tube length =
 30 cm; tube radius = 0.615 cm; discharge energy = 12.5×10^3 joule/mole.

time (ms) f (Hz)	2.70	4.21	5.72	7.20	v _{calc} at T=305°K
2344	399.4	396.3	395.0	394.4	396.6
2734	400.2	396.8	395.7	395.1	397.5
3125	401.1	397.8	396.8	396.9	398.4
3515	402.2	399.2	398.5	398.6	398.7
3906	402.3	399.8	398.9	398.6	399.2
4297	402.4	399.6	398.6	398.0	399.6
4687	401.9	399.2	399.0	-----	399.9

TABLE 19

Sound velocity in m/s as a function of time in excited metastable
 50% He - 50% N₂ mixture at Pressure = 40 Torr; tube length = 40 cm;
 tube radius = 0.615 cm; discharge energy = 16.7×10^3 joule/mole.

Time (ms) f (Hz)	2.91	4.53	6.21	V _{calc} at T=303°K
1953	471.8	468.4	467.2	467.6
2344	471.5	468.6	467.5	469.2
2734	471.3	468.9	468.3	470.4
3125	471.3	469.9	469.3	471.3
3515	471.7	471.1	-----	472.2
3906	472.1	471.2	470.4	472.°
4688	473.8	473.0	471.3	473.5

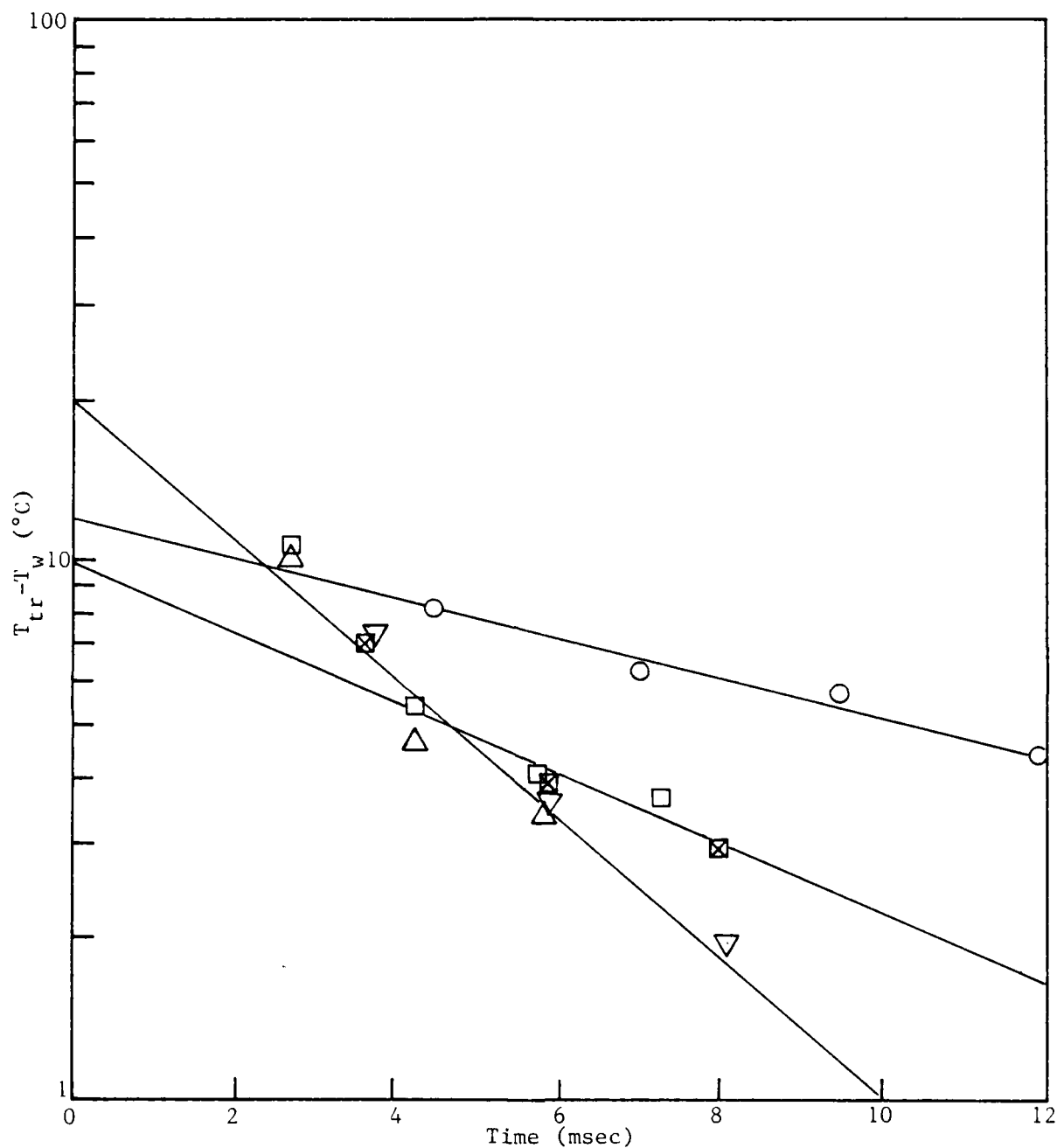


FIGURE 8. The difference between translational and wall temperature ($T_{tr} - T_w$) as a function of time observed in 25% He - 75% N_2 mixture. ΔT_{tr} pressure (P) = 20 Torr; tube radius (R) = 0.615 cm; tube length (L) = 30 cm. ∇ - P = 20 Torr; R = 0.615 cm; L = 43 cm. \square - P = 40 Torr; R = 0.615 cm; L = 30 cm. \otimes - P = 40 Torr; R = 0.615 cm; L = 43 cm. \circ - P = 20 Torr; R = 1.27 cm; L = 50 cm. Solid lines are least squares fit to data.

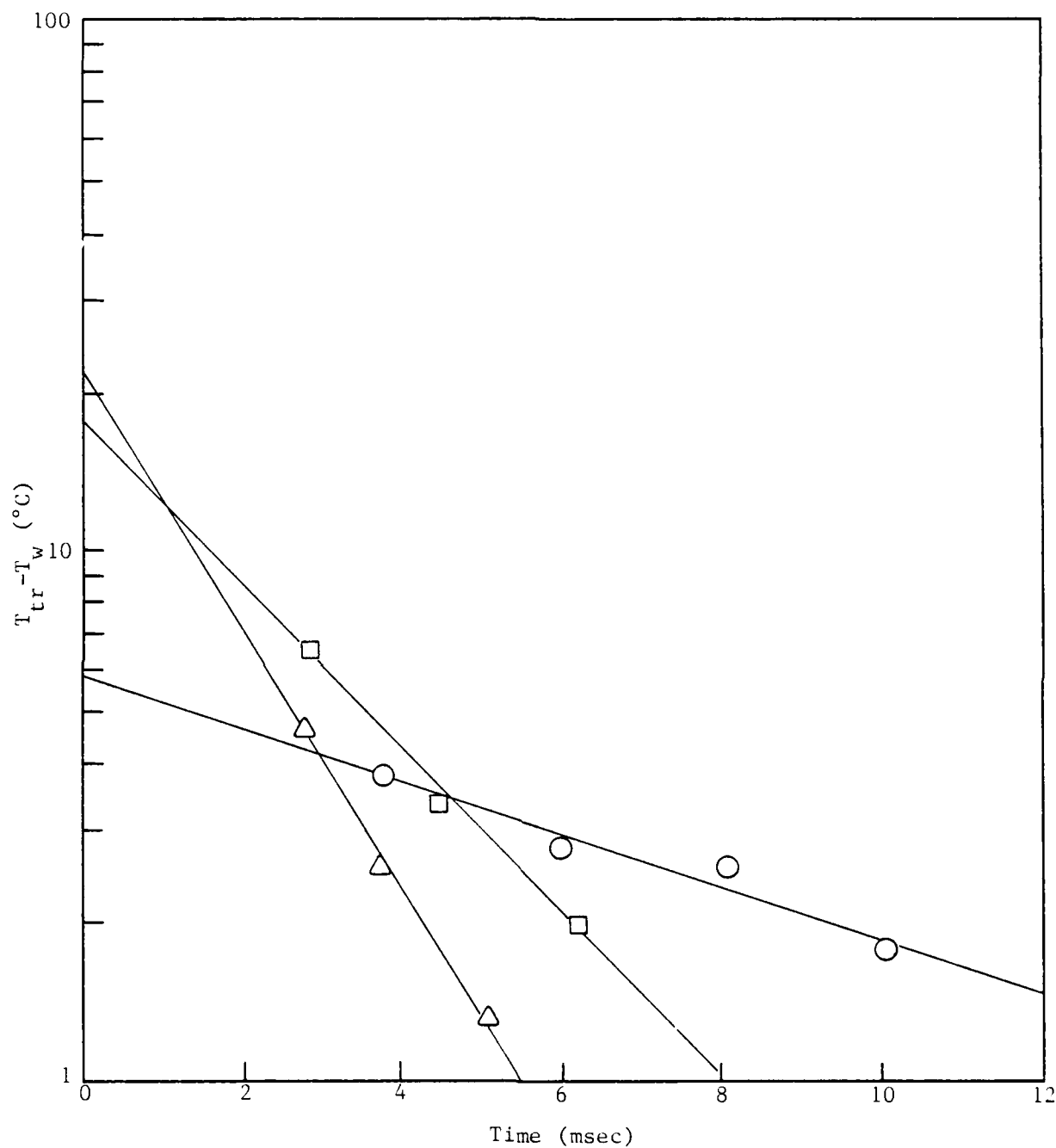


FIGURE 9. The difference between translational and wall temperature ($T_{tr} - T_w$) as a function of time observed in 50% He - 50% N_2 mixture. Δ - $P = 20$ Torr; $R = 0.615$ cm; $L = 30$ cm. \square - $P = 40$ Torr; $R = 0.615$ cm; $L = 40$ cm. \circ - $P = 20$ torr; $R = 1.27$ cm; $L = 60$ cm. Solid lines are least squares fit to data.

TABLE 20

Measured values of the decay time for translational temperature in
 0.615 cm radius tube compared with values of τ_{tc} calculated
 by Equation 19. Values are given in milliseconds.

GAS	P = 20 Torr		P = 40 Torr	
	Observed	Calculated	Observed	Calculated
Pure N ₂	-----	5.78	-----	11.56
25% He - 75% N ₂	3.40	3.58	6.67	7.16
	12.35 ⁽¹⁾	14.26 ⁽¹⁾	-----	-----
50% He - 50% N ₂	1.81	1.89	2.79	3.79
	8.90 ⁽¹⁾	8.06 ⁽¹⁾	-----	-----
10% H ₂ - 90% N ₂	10.22	5.58	35.17	11.20

(1) Tube radius = 1.27 cm.

Results of Measurement in H_2-N_2 Mixtures

In 10% H_2 - 90% N_2 mixtures the vibrational relaxation times are reduced to about 106 and 53 milliseconds at pressures of 20 and 40 Torr respectively.¹⁶ The thermal conduction relaxation times in this mixture at these pressures are approximately 5 and 10 milliseconds for the 1.23 cm tube diameter as predicted by Equation 19. Thus the ratio τ_v/τ_{tc} is relatively small. For a vibrational temperature of 3000°K, the gain, $\Delta\alpha$, is predicted by Shields' calculation to be about 0.009 dB/cm at a pressure of 40 Torr.

The observed time averaged absorption and gain (after subtracting $\Delta\alpha$ observed in pure nitrogen) are presented in Table 21. As expected from Shields' calculations the gain is independent of frequency to the accuracy of measurements. Table 22 and 23 give the observed velocity at 20 and 40 Torr pressures. The corresponding translational temperatures are plotted in Figure 11. As shown in Table 21, the average gain was found to be 0.02 dB/cm at 20 Torr and 0.017 dB/cm at 40 Torr. Figure 10 shows the variations in $\Delta\alpha$ versus time at 40 Torr. At 20 Torr, the time dependent value of $\Delta\alpha$ showed too much scatter to allow a reliable determination of the decay time. This was attributed to the fact that τ_{vc} was shorter than the sound transit time over several reflections.

COMPARISON OF THEORETICAL AND MEASURED RELAXATION TIMES IN THE 10% H_2 - 90% N_2 MIXTURE

When τ_v and τ_{tc} are of comparable sizes the decay time of the translational temperature should be longer than thermal diffusion time

TABLE 21

Time average sound absorption (α) and gain ($\Delta\alpha = \alpha_{\text{calc}} - \alpha_{\text{obs}}$) in excited metastable 10% H_2 - 90% N_2 mixture. Tube radius = 0.615 cm; discharge energy = 44.8×10^3 joule/mole at 20 Torr and 22.4×10^3 joule/mole at 40 Torr. $\Delta\alpha$ has been corrected by observed in pure N_2 . Values are given in dB/cm.

Frequency (Hz)	P = 20 Torr			P = 40 Torr		
	α_{obs}	α_{calc}	$\Delta\alpha$	α_{obs}	α_{calc}	$\Delta\alpha$
1953	0.092	0.119	0.023	0.061	0.084	0.018
2344	0.103	0.130	0.023	0.070	0.092	0.017
2734	0.112	0.141	0.025	0.080	0.099	0.014
3125	0.121	0.150	0.025	0.084	0.106	0.017
3515	0.133	0.159	0.022	0.089	0.112	0.018
3906	0.141	0.168	0.023	0.097	0.118	0.016
4297	0.148	0.176	0.024	0.102	0.124	0.017
4687	0.157	0.183	0.022	0.106	0.129	0.018
5078	0.166	0.191	0.021	0.112	0.134	0.017
5468	0.172	0.198	0.022	0.105	0.139	0.017

TABLE 22

Sound velocity in m/s as a function of time in excited metastable

10% H₂ - 90% N₂ at Pressure = 20 Torr; tube length = 45 cm;

tube radius = 0.615 cm; discharge energy = 48.8×10^3 joule/mole.

time (ms) f (Hz)	3.57	5.70	7.87
1953	-----	369.6	365.9
2344	373.8	370.1	366.6
2734	373.5	370.3	367.1
3125	373.1	370.1	367.8
3515	373.4	369.8	369.2
3906	375.1	370.0	369.9
4297	376.5	371.9	368.4
4687	376.4	372.5	369.2
5078	375.9	372.6	371.7
5468	375.7	372.7	373.8

TABLE 23

Sound velocity in m/s as a function of time in excited metastable

10% H₂ - 90% N₂ at Pressure = 40 Torr; tube length = 40 cm;

tube radius = 0.615 cm; discharge energy = 24.4×10^3 joule/mole.

time (ms) f (Hz)	3.55	5.66	7.80	9.93	12.07
1953	-----	-----	-----	374.0	372.8
2344	380.4	376.9	375.5	374.5	373.6
2734	380.6	377.2	376.2	375.3	374.7
3125	381.1	377.4	377.3	376.6	375.8
3515	382.2	377.8	378.0	377.4	376.0
3906	383.6	378.2	377.7	377.2	376.8
4297	384.3	378.8	377.3	377.5	377.5
4687	384.2	379.6	378.0	377.8	377.8
5076	383.9	380.4	379.3	378.0	377.3
5468	383.7	380.9	379.5	378.3	-----

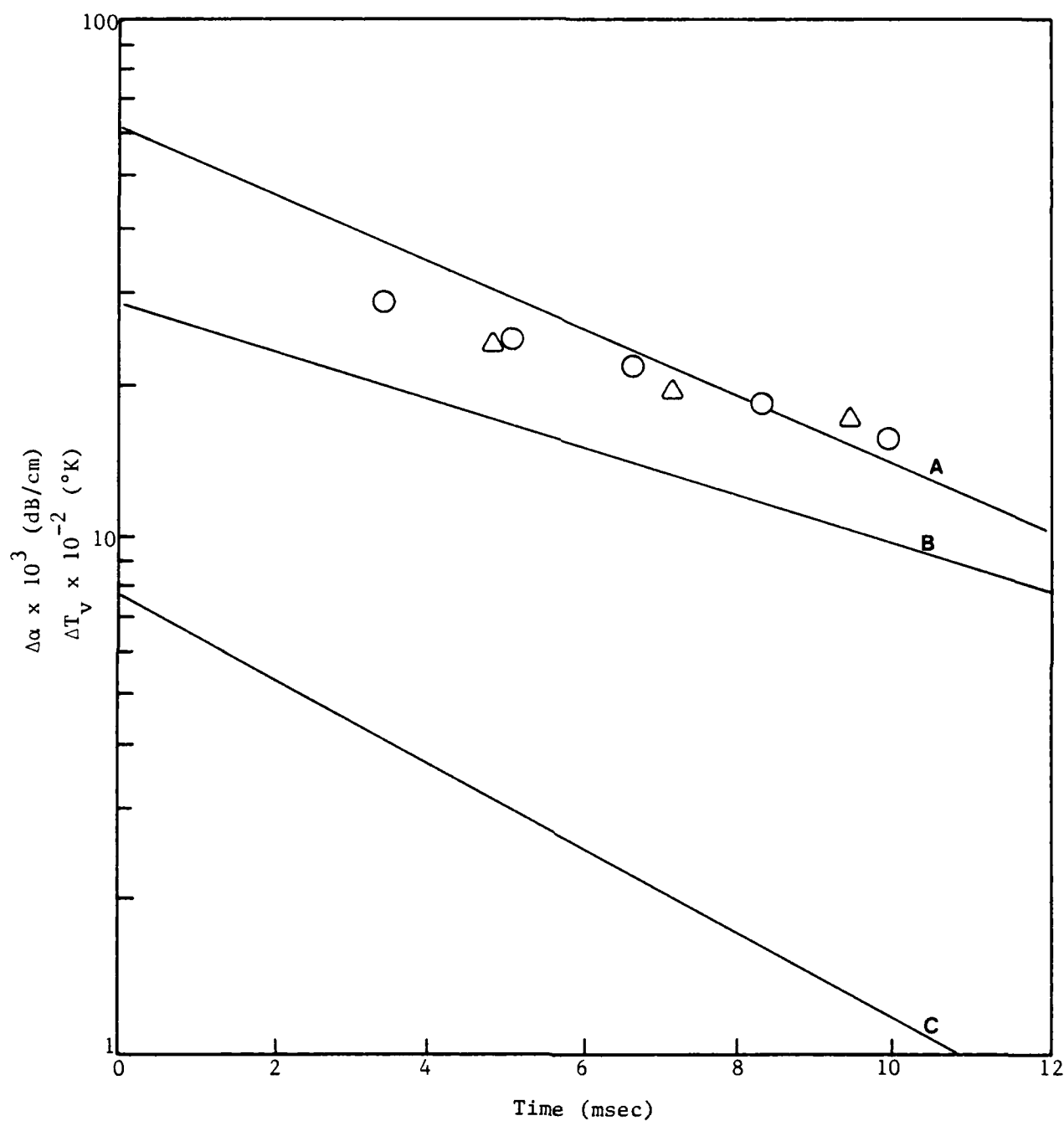


FIGURE 10. Gain ($\Delta\alpha$) as a function of time observed in 10% H_2 - 90% N_2 mixture at pressure = 40 Torr; tube radius = 0.615 cm; discharge energy = 48.8×10^5 joule/mole. Data points, o - observed at tube length = 30 cm; Δ - observed at tube length = 43 cm. Curve A is calculated gain setting $d(\ln T_v^{-1})/dT = 0.05$. Curve B is $\Delta T_v \times 10^{-2}$ computed from Equation 37. Curve C is the calculated gain by setting $d(\ln T_v^{-1})/dT = 0.01$.

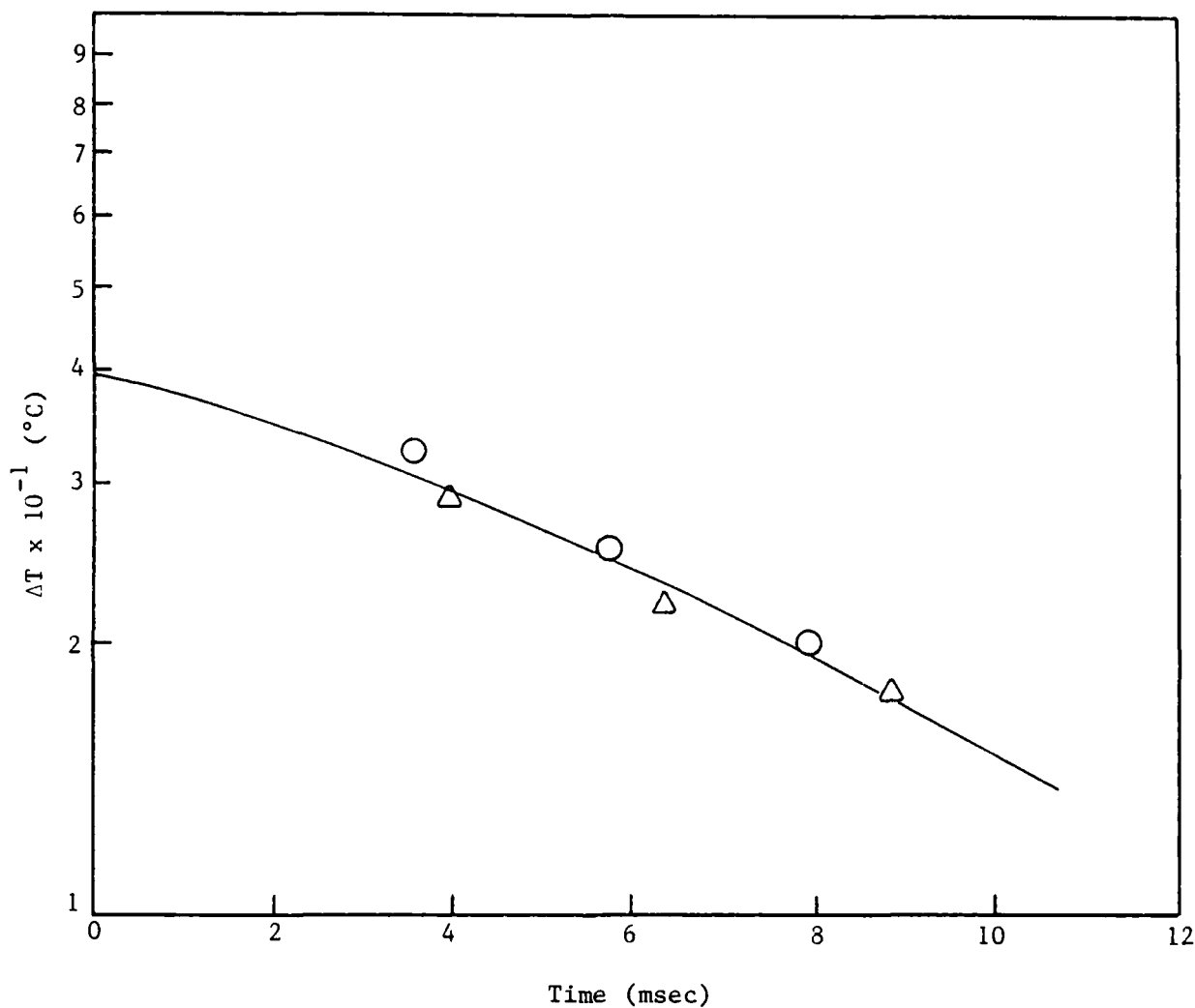


FIGURE 11. The difference between translational and wall temperature (ΔT) as a function of time for experiment on 10% H_2 - 90% N_2 mixture at pressure = 20 Torr; tube radius = 0.615 cm; discharge energy = 48.8×10^3 joule/mole. Data points, O - observed at tube length = 40 cm; Δ - observed at tube length = 45 cm. Solid curve is the theoretical curve computed from Equation 34.

due to the feeding of energy into translation from vibration. The difference is noted between the measured and calculated relaxation times for 10% H_2 - 90% N_2 mixtures in Table 20. The energy going from vibration to translation also makes the decay time for vibrational temperature shorter than the calculated τ_{tc} . The relationship between τ_{tc} as calculated from Equation 19 and these decay times for translational and vibrational temperature as determined from measured velocities can be obtained as follows. Assume the translational mode gains energy from the vibrational mode and loses it to the wall. The time rate of change of the translational and vibrational energy can be written as

$$\frac{dE_{tr}}{dt} = \frac{C_i(T_v)}{\tau_v} (\tau_v - T) - \frac{C_\infty}{\tau_{tc}} (T - T_w), \quad (25)$$

and

$$\frac{dE_v}{dt} = - \frac{C_i(T_v)}{\tau_v} (T_v - T) - \frac{C_i(T_v)}{\tau_{vc}} (T_v - T_w), \quad (26)$$

where E_{tr} is the translational energy, E_v is the vibrational energy, T_v , T , and T_w are the vibrational, translational, and wall temperature respectively, C_i is the vibrational specific heat, and C_∞ is the time independent specific heat equal to $5/2 R$ per mole for N_2 and H_2 . τ_v , τ_{vc} and τ_{tc} are the same as defined before. By setting

$$T_v - T = (T_v - T_w) - (T - T_w), \quad (27)$$

and

$$\frac{dE_{tr}}{dt} = \frac{C_\infty dT}{dT}; \quad \frac{dE_v}{dT} = \frac{C_i dT_v}{dT} \quad (28)$$

and assuming exponential time dependence the differential equations 25 and 26 are reduced to algebraic equations:

$$\left[\lambda - \left(\frac{C_i}{C_\infty \tau_v} + \frac{1}{\tau_{tc}} \right) \right] \Delta T + \frac{C_i}{C_\infty \tau_v} \Delta T_v = 0 \quad (29)$$

$$\frac{1}{\tau_v} \Delta T + \left\{ \lambda - \frac{1}{\tau_v} - \frac{1}{\tau_{vc}} \right\} \Delta T_v = 0 \quad (30)$$

where $\Delta T = T - T_w$ and $\Delta T_v = T_v - T_w$. The solution of the secular equation gives two permissible values of λ in terms of the known relaxation times,

$$\lambda_1 = \frac{1}{\tau_v} \left(\frac{C_i}{C_\infty} + 1 \right) + \frac{1}{\tau_{tc}} \quad (31)$$

$$\lambda_2 = \frac{1}{\tau_{tc}}. \quad (32)$$

The solutions for ΔT and ΔT_v are then

$$\Delta T = A_1 e^{-\lambda_1 t} + A_2 e^{-\lambda_2 t} \quad (33)$$

$$\Delta T_v = A_3 e^{-\lambda_1 t} + A_4 e^{-\lambda_2 t} \quad (34)$$

where A_1 and A_2 can be determined from the measured values for ΔT and $\frac{d(\ln \Delta T)}{dt}$ at a particular time. For this purpose we choose ΔT values from Figures 11 and 12 corresponding to t approximately equal to τ_{tc} . This value of time was chosen since for $t < \tau_{tc}$, the excited metastable state had no time to develop and the gas temperature was not uniform in the tube.

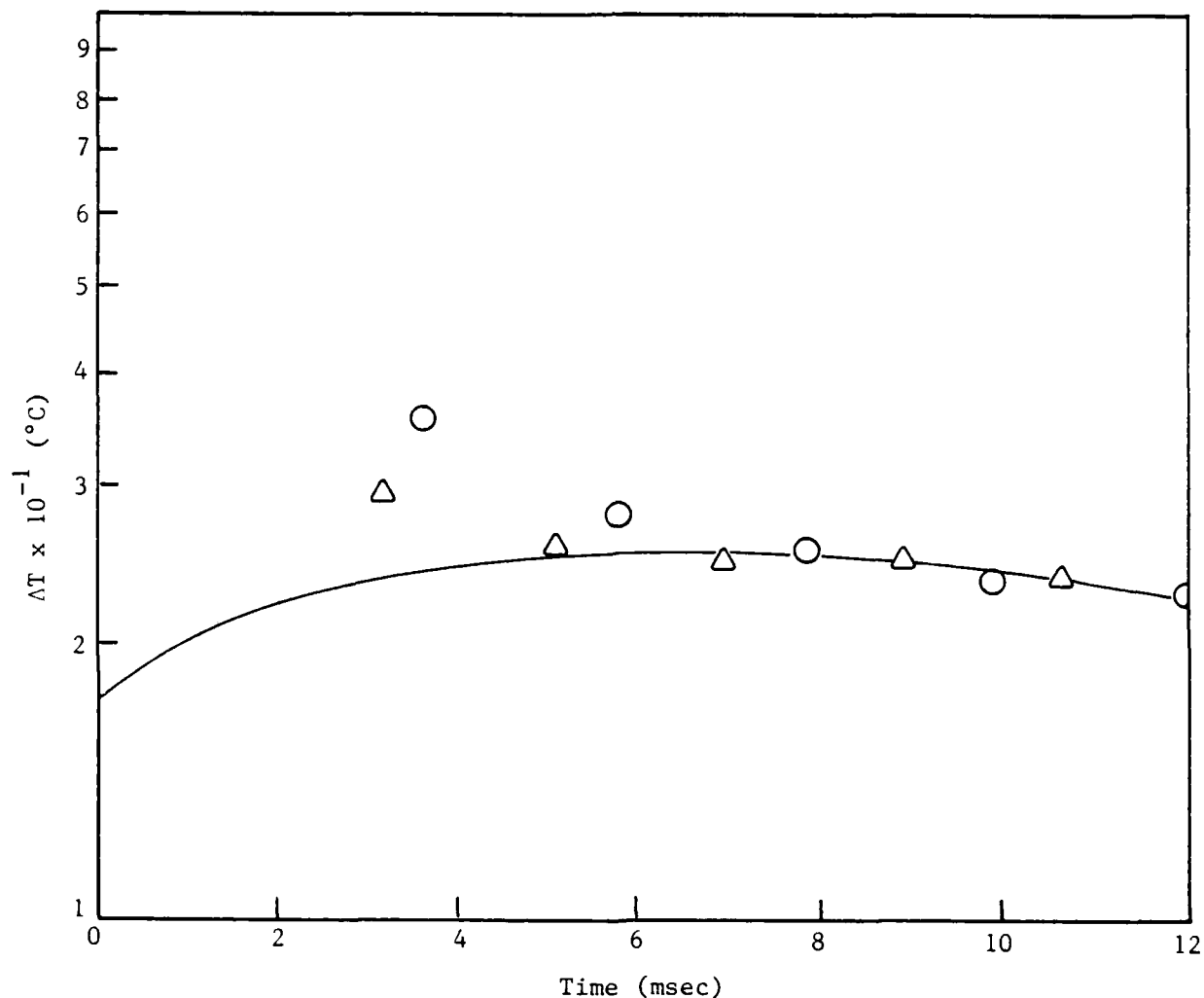


FIGURE 12. The difference between translational and wall temperature (ΔT) as a function of time for experiment on 10% H_2 - 90% N_2 mixture at pressure = 40 Torr; tube radius = 0.615 cm; discharge energy = 24.4×10^3 joule/mole. Data points, Δ - observed at tube length = 35 cm; \circ - observed at tube length = 40 cm. Solid curve is the theoretical curve computed from Equation 39.

A_3 and A_4 can be related to A_1 and A_2 by substituting Equations 33 and 34 into Equations 25 and 26. Separating the terms containing $\exp(-\lambda_1 t)$ and those containing $\exp(-\lambda_2 t)$ one obtains

$$A_3 = -\frac{C_\infty}{C_i} A_1 \quad (35)$$

and

$$A_4 = A_2. \quad (36)$$

The procedure in analyzing the 10% H_2 - 90% N_2 data then is as follows

1. Assume an average value for the vibrational temperature, T_v , over the period of measurement, and determine C_i from C_v^{24} using the equation $C_i = C_v - C_\infty$.
2. Using known values of τ_v , and τ_{tc} ($=\tau_{vc}$), and the assumed C_i determine λ_1 and λ_2 from Equations 31 and 32.
3. From the values and slopes of curves in Figures 11 and 12 at $\tau \approx \tau_{tc}$, determine A_1 and A_2 , A_3 and A_4 as explained above.
4. Calculate ΔT_v using Equation 34 over the measured time interval and compare with the average value assumed in Step 1.
5. Repeat until the values of ΔT_v obtained in Step 4 are consistent with values of T_v assumed in Step 1.

Using a value of $C_i = 0.21R$ corresponding to $T_v = 1500^\circ K$; values of $\Delta T = 24^\circ$, and $[\frac{d}{dt}(\ln T)]^{-1} = 9.05$ milliseconds corresponding to values

from Figure 11 at $t = 6$ milliseconds; calculating τ_{tc} from Equation 19; setting $\tau_{vc} = \tau_{tc}$; and obtaining τ_v from Reference 16 we obtained

$$\Delta T(^{\circ}\text{C}) = 532.5e^{-0.179t} - 492e^{-0.190t} \quad (37)$$

and

$$\Delta T_v(^{\circ}\text{C}) = 532.5e^{-0.179t} + 4731.4e^{-0.190t} \quad (38)$$

where t is in milliseconds. The curve in Figure 11 drawn through the points taken at 20 Torr is calculated from Equation 37. In a similar way the equations

$$\Delta T(^{\circ}\text{C}) = 238.0e^{-0.089t} - 220.3e^{-0.110t} \quad (39)$$

and

$$\Delta T_v(^{\circ}\text{C}) = 238.0e^{-0.089t} + 2622.4e^{-0.110t} \quad (40)$$

fit data points observed at 40 Torr pressure in Figure 12. The constants in the 40 Torr equation were obtained by using values of $\Delta T = 24^{\circ}\text{C}$ and $d(\frac{\ln \Delta T}{dt})^{-1} = 37.84$ msec as determined from Figure 12 at $t = 10$ msec and by setting $C_i = 0.21R$ corresponding to $T_v = 1500^{\circ}\text{K}$. It should be noted that these equations have been obtained from the velocity measurement without reference to absorption.

The agreement between the measured and calculated time dependence of ΔT in Figure 11 is impressive since the only adjustable parameter is the vibrational temperature.

In Figure 12 the time dependence is correctly predicted after approximately 10 msec, but the velocity obtained from the first reflected pulse is too high. This was also the case for the 50% He - 50% N₂ mixture at 40 Torr. Here, as there, the difference can be attributed to the fact that the first reflection is occurring for times shorter than τ_{tc} and the temperature variations left by the discharge have not been smoothed out nor the metastable state developed.

Equations 38 and 40 give the vibrational temperature, ΔT_v , as a function of time. Note these equations have been determined just from measured ΔT vs time values and known values of τ_v , τ_{tc} and τ_{vc} . The value of ΔT_v predicted at $t = \tau_{tc}$ is 2100°K for 20 Torr and 1200°K for 40 Torr. These values would imply that 8.4% of the discharge energy was in the gas at that time. This percentage is unreasonably small. The gain measurements in the next section indicate a higher vibrational temperature was present. This discrepancy would infer that the value of τ_v in H₂-N₂ mixtures used in calculating the constants in Equation 39 and 40 which were obtained from fluorescent measurements by Frev, et al.¹⁶ were too small.

COMPARISON OF THEORETICAL AND MEASURED GAIN IN 10% H₂ - 90% N₂ AND 50% He - 50% N₂.

In two cases it has been possible to plot the gain versus time (50% He - 50% N₂ at 40 torr and 10% H₂ - 90% N₂ at 40 Torr). For the H₂-N₂ mixture it has been possible to get T_v vs time from the velocity measurements as well. It is now interesting to compare these measured gains with values calculated from Shields' theory. For 10% H₂ - 90% N₂, the

measured values of gain vs time are plotted in Figure 10 also shown plotted in this figure to a different scale is ΔT_v vs time calculated by Equation 40 (curve B). Since both T_v and T are determined by Equations 39 and 40 as a function of time, it is also possible to calculate expected gain versus time using Shields' theory. The result is plotted in Figure 10 as curve C assuming value $\frac{d}{dT}(\ln \tau_v^{-1}) = 0.01$. The result gives magnitudes of gain that are too small by a factor of 10 and a negative slope that is too large by a factor of two. There are three parameters that affect the calculated gain: τ_v , $\frac{d}{dT}(\ln \tau_v^{-1})$, and T_v . The magnitude of the gain can be increased by increasing $\frac{d}{dT}(\ln \tau_v^{-1})$ or T_v , or decreasing τ_v . However the slope of the gain with time is changed little by changing any of these. The measured small decrease in gain with time implies a T_v that changes more slowly with time than predicted by Equation 40. This discrepancy may be due to the fact that most of the measurements in this mixture were made for times less than τ_{tc} while the temperature T_v and T were still probably far from uniform in the tube. The appropriate magnitude of the gain would be predicted by Shields' calculation if 70% of the discharge were in vibration at $t = \tau_{tc}$ msec and $\frac{d}{dT}(\ln \tau_v^{-1}) = 0.05$. Curve A in Figure 10 corresponds to values calculated by Shields' theory assuming $\frac{d}{dT}(\ln \tau_v^{-1}) = 0.05$ and T_v equal to 3000°K at $t = 7.5$ msec which decays with decay time equal to τ_{tc} . This would correspond to 70% of the discharge energy residing in the vibrational energy of the gas at $t \approx \tau_{tc}$.

For the case of 50% He - 50% N₂ mixture at 40 Torr, it was not possible to get T_v vs time from the velocity measurements. However, because τ_v is so much larger than τ_{tc} in this gas it is safe to assume T_v decays with a relaxation time equal to τ_{tc} . Assuming the same percentage of discharge energy goes into N₂ vibration in this mixture as with the H₂ mixture, a value of T_v at $t = \tau_{tc}$ would equal 6000°K. Using this value of T_v and again assuming $\frac{d}{dt} \ln(\tau_v^{-1}) = 0.05$. The calculated gain in 50% He = 50% N₂ is plotted as a solid line in Figure 7.

AN EXAMINATION OF THE CONSISTENCY OF THE RESULTS.

It is interesting to note a number of consistencies in the data. First consider the ΔT vs time data that has been obtained from the velocity measurements.

1. The initial translational temperatures that result from the discharge are reasonable. In pure N₂ the translational temperature changed only slightly as expected due to the small amount of discharge energy going directly into translation and the big value of τ_v/τ_{tc} . In the mixtures, the value of ΔT_0 increase approximately linearly with the discharge energy as might be expected (see Figure 13).

2. The time dependence of ΔT in all mixtures agrees roughly with values calculated from values of τ_{tc} , τ_{vc} and τ_v obtained independently (Table 20).

3. The gain per unit length was found to be independent of frequency as predicted theoretically for the frequency range considered (see Table 21).

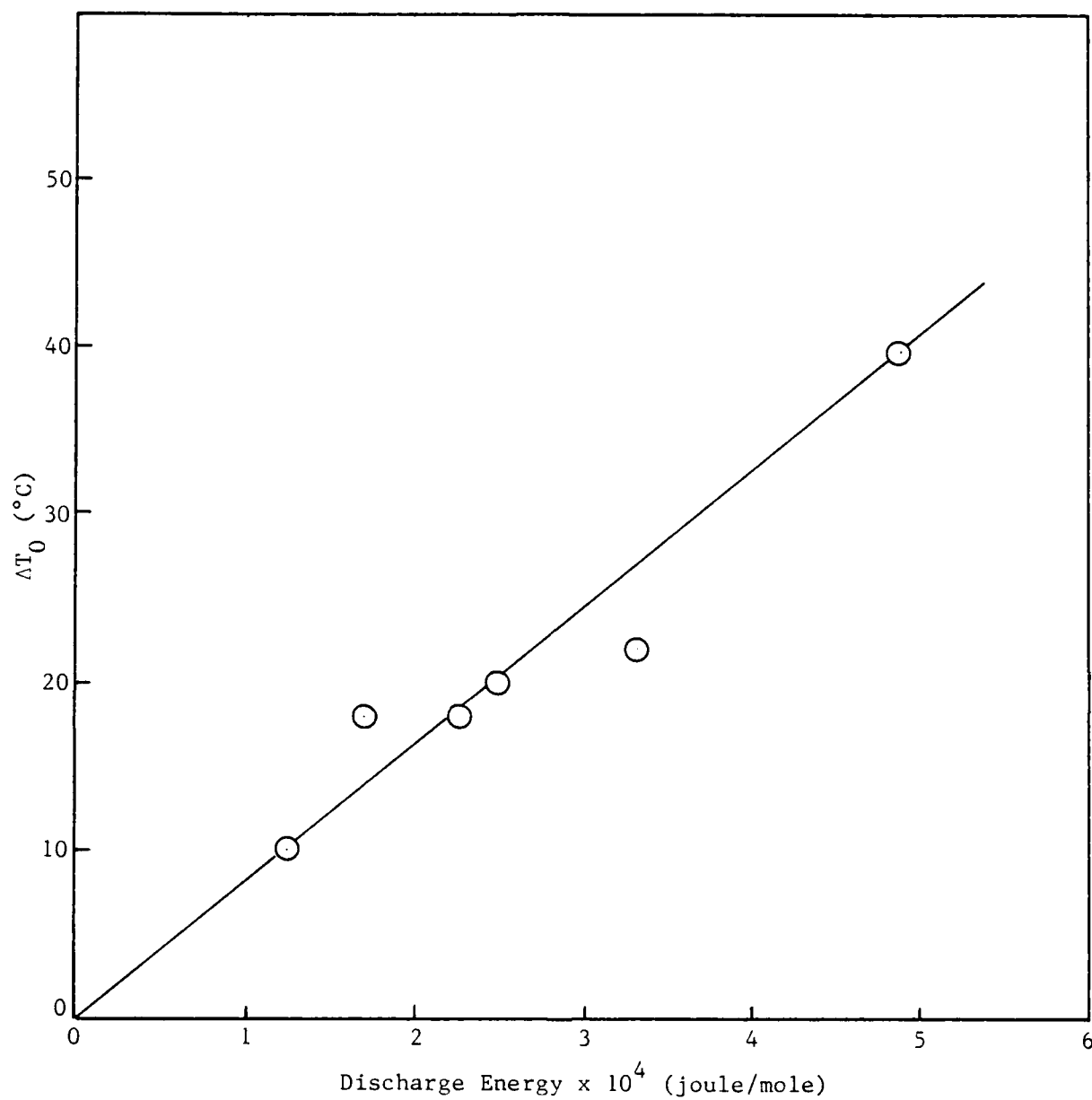


FIGURE 13. The increase in translational temperature (ΔT_0) as a function of discharge energy in gases under study. Solid line is visual straight line fit to data.

4. In both 10% H_2 - 90% N_2 and 50% He - 50% N_2 , the gain vs time curve indicates the temperature dependence of the transition rate $\frac{d}{dT}(\tau_v^{-1})$ is given by $\frac{d}{dT}\ln(\tau_v^{-1}) = 0.05$ and 70% of discharge energy is in the vibrational mode of the gas.

There are two aspects of the data still unexplained:

1. The measured gain is larger than predicted by Shields' theoretical calculation.

2. In 10% H_2 - 90% N_2 the values of ΔT_v calculated from the time dependence of the ΔT data and known value of τ_{tc} , τ_{vc} and τ_v are unreasonable when compared with values estimated from the amount of discharge energy going into vibration.

CHAPTER V

CONCLUSIONS

An investigation to observe sound amplification in a vibrationally excited gas has been described. By means of an electrical discharge, nitrogen gas was vibrationally excited to a vibrational temperature of the order of a few thousand degrees Kelvin. The discharge was also able to generate a sound pulse that propagated through the excited gas after the discharge had ceased. By discrete Fourier analysis of the sound pulse upon each reflection in the discharge tube it was possible to measure the absorption and velocity for different frequencies as a function of time. By comparing the measured velocity with the numerically calculated velocity, the variation in translational temperature of the gas with time could be determined. In some cases (50% He - 50% N₂ at 20 Torr, 50% He - 50% N₂ at 40 torr and 10% H₂ - 90% N₂) measureble gain was observed. In two cases (50% He - 50% N₂ at 40 torr and 10% H₂ - 90% N₂ at 40 Torr) it was possible to measure the variation of gain with time.

The measured decay rate for the difference between translational temperature and wall temperature (ΔT) agreed in all gas mixtures and pressures with values calculated from independently determined values of the vibrational relaxation time, τ_v , and the thermal diffusion time, τ_{tc} , assuming that the thermal diffusion time for conducting translational energy to the wall was the same as that for conducting vibrational energy to the wall ($\tau_{tc} = \tau_{vc}$).

Experiments on pure nitrogen confirmed Nighan's theoretical calculation¹⁵ from measured electron cross section that practically all the discharge energy goes into vibration in N_2 , i.e., the discharge was found to raise the translational temperature in pure N_2 only a few degrees. In the gas mixtures, the discharge was found to raise the translational temperature from 10 to 40°C depending upon the energy per mole deposited.

In those cases where the gain was measured it was about an order of magnitude greater than expected from Shields' calculations. These calculations were based on the assumption that the temperature dependence of the transition rate was given by $\frac{d}{dT} \ln(\tau_v^{-1}) = 0.01$. By increasing this derivative to 0.05 and assuming 70% of the discharge energy was in vibration at $t \approx \tau_{tc}$, Shields' calculations would give the approximate magnitude of gain in both the gases where this time dependence was measured (50% He - 50% N_2 and 10% H_2 - 90% N_2 at 40 Torr).

In the 10% H_2 - 90% N_2 gas at 40 Torr, τ_v/τ_{tc} was small enough so that it was possible to imply the ΔT_v vs time curve from ΔT versus time curve. However ΔT_v values thus obtained indicate that only about 8% of the energy stored in the capacitor was coupled into the vibrational energy of the gas.

The time averaged gain in dB/cm over a frequency range from 1.9 to 5.5 kHz was found, within the accuracy of measurement, to be independent of frequency as predicted by Shields' calculations.

Measurements were also tried in a 20% H_2 - 80% N_2 mixture at a pressure of 40 Torr. The signal pattern was unreproducible. It was

observed that even though the first reflections of the sound pulse for different sparks coincided, the following reflections were randomly shifted along the time axis (on the oscilloscope screen). This meant that the velocity (and thus the gas temperature) was not varying with time in the same way from spark to spark. This was attributed to the small value of the ratio τ_v/τ_{tc} . Evidently the gas was being heated unevenly by the vibrational energy relaxing into translation faster than it could be conducted to the wall. Due to these difficulties the experimental data for 20% H_2 - 80% N_2 mixture was not reported in this study. However, qualitatively, the gain was observed in this mixture to have a slightly larger value than the gain observed in 10% H_2 - 90% N_2 mixture.

The results reported here suggest two definitive experiments. (1) Make measurements on mixture of N_2 and H_2O . The H_2O will shorten the relaxation time but should produce less gain since the vibrational relaxation rate is nearly temperature independent. (2) A second experiment suggested is on mixtures of CO and H_2 . CO like N_2 should be left highly vibrationally excited by the discharge. In this case the vibrational temperature of the CO can be monitored by the infrared emission from vibrational transition.

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APPENDIX A

Computer program used to fourier analyze the sound pulse and calculate the absorption and velocity.

```

10 OPTION BASE 1
20 SHORT V(256),R(256),I(256)
30 SHORT H(30,7),A(30),C(30,7),T(30),H1(30,7),T1(30,7)
40 SHORT S1(7),S2(7),T0(30,7),S3(7),S4(7),A1(30),C1(30,7)
50 DIM AS[8200],BS[280]
60 DISP "GAS SAMPLE"
70 INPUT GS
80 DISP "INPUT P,L,T9"
90 INPUT P9,L9,T9
100 PRINT "GAS SAMPLE=";GS
110 PRINT "PRESSURE=";P9;"MM"
120 PRINT "L TUBE=";L9;"CM"
130 PRINT "R.TEMP.=";T9;"DEG.C"
140 DISP "# OF POINTS N, # OF REFLECTION M1, FREQ. COMP M2, # OF SPARK
M3"
150 INPUT N,M1,M2,M3
160 PRINT "# OF SAMPLE POINTS=";N
170 T=N*.00001
180 FOR M=1 TO M1
190 FOR I=1 TO M2
200 H1(I,M)=0 @ T1(I,M)=0 @ C1(I,M)=0
210 S3(M)=0 @ S4(M)=0 @ A1(I)=0
220 NEXT I @ NEXT M
230 FOR K9=1 TO M3
240 PRINT @ PRINT "SPARK #",K9
250 FOR M=1 TO M1
260 DISP "INPUT S1(";M;"),S2(";M;")"
270 INPUT S1(M),S2(M)
280 S3(M)=S3(M)+S1(M)
290 S4(M)=S4(M)+S2(M)
300 PRINT "REFL #";M;S1(M);S2(M)
310 NEXT M
320 FOR M=1 TO M1
330 GOSUB 1480
340 FOR I=S2(M)-S1(M)+2 TO N
350 V(I)=0
360 NEXT I
370 FOR I=1 TO N/2

```

```

380 IF 2*I-1=1 THEN 440
390 IF 2*I=S2(M)-S1(M)+1 THEN 440
400 IF 2*I=S2(M)-S1(M)+1 THEN 460
410 R(I)=V(I*2-1)
420 I(I)=V(I*2)
430 GOTO 470
440 R(I)=.5*V(2*I-1)
450 GOTO 420
460 I(I)=.5*V(2*I) @ R(I)=V(2*I-1)
470 NEXT I
480 P=1
490 FOR I=1 TO 10
500 P=P*2
510 IF P=N THEN 530
520 NEXT I
530 P1=I
540 N2=N/2
550 J=0
560 GOSUB 1100
570 FOR I=1 TO M2
580 TO(I,M)=ATN2(I(I),R(I))-2*PI*(I-1)*S1(M)/N
590 H(I,M)=SQR(I(I)^2+R(I)^2)
600 T1(I,M)=T1(I,M)+TO(I,M)
610 H1(I,M)=H1(I,M)+H(I,M)
620 NEXT I
630 BEEP @ NEXT M
640 GOSUB 1670
650 FOR I=1 TO M2
660 A1(I)=A1(I)+A(I) @ NEXT I
670 GOSUB 1850
680 FOR M=1 TO M1-1
690 FOR I=1 TO M2
700 C1(I,M)=A1(I,M)+C(I,M)
710 NEXT I @ NEXT M
720 IF K9=M3 THEN 750
730 BEEP @ BEEP @ DISP "INPUT SPARK#";K9+1 @ PAUSE
740 NEXT K9
750 BEEP @ BEEP @ BEEP @ DISP "DONE!!!"
760 FOR I=1 TO M2
770 A1(I)=A1(I)/M3
780 PRINT I-1;A1(I) @ NEXT I
790 FOR M=1 TO M1-1
800 PRINT @ PRINT "VEL. OF REFL.#";M;
810 FOR I=1 TO M2
820 C1(I,M)=C1(I,M)/M3
830 PRINT I-1;C1(I,M)
840 NEXT I @ NEXT M
850 FOR M=1 TO M1
860 S3=S3(M)/M3

```

```

870 S4=S4(M)/M3
880 PRINT @ PRINT "REFL.#";M;"STARTS AT";S3;"ENDS AT";S4
890 FOR I=1 TO M2
900 H(I,M)=H1(I,M)/M3
910 TO(I,M)=T1(I,M)/M3
920 PRINT USING 930; I-1;H(I,M);TO(I,M)
930 IMAGE 40,2X,MZ,3DE,2X,MZ,3DE
940 NEXT I
950 NEXT M
960 PRINT @ PRINT "SLOPES OF LOG(H) VS,M"
970 GOSUB 1670
980 FOR I=1 TO M2
990 PRINT I-1;A(I) @ NEXT I
1000 GOSUB 1850
1010 FOR M=1 TO M1-1
1020 PRINT "VEL. OF REFL.#";M
1030 FOR I=1 TO M2
1040 PRINT I-1;C(I,M)
1050 NEXT I @ NEXT M
1060 BEEP @ BEEP @ END
1070 REM
1080 REM SUBROUTINE 1070 TO 1410 FOURIER ANALYZES DATA POINTS
1090 REM
1100 RAD
1110 K=0
1120 FOR J=1 TO N2-1
1130 I=2
1140 IF K<N2/I THEN 1160
1150 K=K-N2/I @ I=I+I @ GOTO 1140
1160 K=K+N2/I
1170 IF K<=J THEN 1190
1180 A=R(J+1) @ R(J+1)=R(K+1) @ R(K+1)=A @ R=I(J+1) @ I(J+1)=I(K+1) @
I(K+1)=A
1190 NEXT J
1200 G=.5 @ P2=1
1210 FOR I=1 TO PI=1
1220 G=G+G @ C=1 @ E=0 @ O=SQR((1=P2)/2)
1230 P2=(1-2*(I-1))*SQR(1+P2)/2)
1240 FOR R=1 TO G
1250 FOR J=R TO N2 STEP G+G
1260 K=J+G @ A=C*R(K)+E*I(K) @ R=E*R(K)-C*I(K) @ R(K)=R(J)-A @
I(K)=I(J)+B @ R(J)=R(J)+A @ I(J)=I(J)-B
1270 NEXT J
1280 A=F*P2+C*Q @ C=C*P2-E*O @ E=A
1290 NEXT R
1300 NEXT I
1310 A=PI/N2 @ P2=COS(A) @ O=SIN(A) @ A=R(1) @ R(1)=A+I(1) @ I(1)=A-I(1)
1320 R(1)=R(1)/2 @ I(1)=I(1)/2
1330 C=1 @ E=0

```

```

1340 FOR J=2 TO N2/2
1350 A=E*P2+C*Q @ C=C*P2-E*Q @ E=A @ K=N2-J+2 @ A=R(J)+R(K)
1360 B=(I(J)+I(K))*C-(R(J)-R(K))*E @ U=I(J)-I(K)
1370 V=(I(J)+I(K))*E+(R(J)-R(K))*C
1380 R(J)=(A+B)/2 @ I(J)=(U-V)/2 @ R(K)=(A-B)/2 @ I(K)=-((U+V)/2)
1390 NEXT J
1400 I(N2/2+1)=-I(N2/2+1)
1410 FOR J=1 TO N2
1420 R(J)=R(J)/N2 @ I(J)=I(J)/N2
1430 NEXT J
1440 RETURN
1450 REM
1460 REM SUBROUTINE 1420 TO 1560 TRANSFERS DATA POINTS FROM NICOLET TO
    COMPUTER
1470 REM
1480 OUTPUT 715; "NO"
1490 IOBUFFER BS
1500 TRANSFER 714 TO BS FHS; COUNT 230
1510 CLEAR 7
1520 VO=VAL(BS[5,8])
1530 V1=VAL(BS[14,20])
1540 OUTPUT 715; "D3D2"
1550 IOBUFFER AS
1560 TRANSFER 714 TO AS FHS; COUNT 8192
1570 CLEAR 7
1580 J=0
1590 FOR I=S1(M)*2+1 TO S2(M)*2+1 STEP 2
1600 J=J+1
1610 V(J)=V1*(256*NUM(AS[I])+NUM(AS[I+1]))-65536*(NUM(AS[I])>=128))
1620 NEXT I
1630 RETURN
1640 REM
1650 REM SUBROUTINE 1580 TO 1720 CALCULATES THE SLOPE OF LOG(AMPL.) VS.
    THE # OF REFLECTION
1660 REM
1670 FOR I=1 TO M2
1680 X0=0 @ X1=0 @ Y0=0 @ S=0
1690 FOR M=1 TO M1
1700 Y=LOG(H(I,M))
1710 X=M
1720 X2=X^2 @ S1=X*Y
1730 X0=X0+X @ X1=X1+X2
1740 Y0=Y0+Y @ S=S+S1
1750 NEXT M
1760 D=X1*M1-X0^2
1770 D1=S*M1-X0*Y0
1780 D2=X1*Y0-S*X0
1790 A(I)=D1/D
1800 NEXT I

```



```
1810 RETURN
1820 REM
1830 REM SUBROUTINE 1730 TO 1800 CALCULATES VELOCITY
1840 REM
1850 FOR M=1 TO M1-1
1860 FOR I=1 TO M2
1870 J=I-1
1880 T(I)=TO(I,M+1)-TO(I,M)
1890 C(I,M)=- (2*PI*J*L9/T/T(I))
1900 NEXT I
1910 NEXT M
1920 RETURN
```

APPENDIX B

TABLE 24

Physical Properties of gases study at 300°K used in calculations.

Gas	Molecular Weight	$\frac{C_v}{R}$	Thermal	
			Conductivity (cal cm ⁻¹ deg ⁻¹ sec ⁻¹)	Viscosity (gr cm ⁻¹ sec ⁻¹)
N ₂	28.013	2.5	602.6x10 ⁻⁷	1.7848x10 ⁻⁴
25He-75N ₂	22.011	2.25	869.2x10 ⁻⁷	1.8542x10 ⁻⁴
50He-50N ₂	16.008	2.00	1470x10 ⁻⁷	1.9370x10 ⁻⁴
10H ₂ -90N ₂	26.828	2.50	623.6x10 ⁻⁷	1.7649x10 ⁻⁴

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SOME AMPLIFICATION BY RELAXATION PROCESSES IN WINGS
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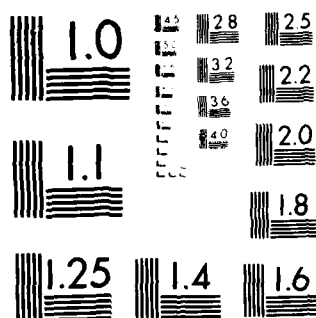
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MICROCOPY RESOLUTION TEST CHART
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BIOGRAPHICAL SKETCH OF THE AUTHOR

Dadang Iskandar, the author, was born in Ciamis, West Java, Indonesia on July 6, 1941 the son of St. Marah and M. Halil; graduated from High School, Yogyakarta, Central Java in 1961; received a Bachelor of Science degree from the University of Gajah Mada, Yogyakarta in November 1967; received a Master of Science degree from the University of Mississippi in May 1982.

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